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manual of food quality control

16. radionuclides in food

Food
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16. radionuclides in food

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FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED NATIONS
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PREFACE

The world's attitude toward radionuclide contamination of foods suddenly changed on April 26, 1986. On that date the accident and fire at the nuclear reactor at the Chernobyl power station released a considerable amount of radioactive substances into the atmosphere. Radioactive particles were transported over Europe and Asia by air currents and eventually settled to earth to begin a new life as environmental contaminants. Widespread monitoring of radionuclide contamination of foods had been commonplace in the 1950's and 60's. However, since the cessation of above-ground testing of nuclear weapons in the 1960's there had been little public concern and consequently reduced monitoring of radionuclides in foods.

As a result of the widespread fallout of radionuclides from Chernobyl, nations began intensive sampling and analysis efforts to determine the level and extent of the contamination. Food sampling and regular monitoring of radioactivity levels were activated by all the countries affected by the fallout and by countries importing food from affected areas.

In December 1986 FAO convened an Expert Consultation in Rome to discuss and recommend limits for radionuclide contamination of foods. As the result of this consultation and further elaboration within the Codex Alimentarius Commission, Guideline Levels for Radionuclides in Foods Following Accidental Nuclear Contamination for Use in International Trade were established. These Guidelines are published in Codex Alimentarius, Volume 1, FAO and WHO, 1992, ISBN 92-5-103120-7.

Although years have passed since the Chernobyl accident, nations continue to carry out monitoring activities for radionuclides. In an effort to provide assistance and standardization to public and private radionuclide monitoring systems, the Food and Agriculture Organization of the United Nations has prepared this comprehensive manual which contains background information along with sampling and analysis methodology for determining radionuclides in foods.

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This publication is available to persons and organizations. Comments and suggestions for possible further editions of this publication should be send to:

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SPECIAL NOTE

The Laboratory procedures described in this Manual are designed to be carried out by properly trained personnel in a suitably equipped laboratory. In common with many such procedures, they may involve hazardous materials.

For the correct and safe execution of these procedures, it is essential that laboratory personnel follow standard safety precautions.

While the greatest care has been exercised in the preparation of this information, FAO expressly disclaims any liability to users of these procedures for consequential damages of any kind arising out of or connected with their use.

The analytical procedures detailed herein are also not to be regarded as official because of their inclusion in this Manual. They are simply procedures which have been found to be accurate and reproducible in a variety of laboratories.

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RADIONUCLIDES IN FOOD

I. INTRODUCTION TO RADIOACTIVITY & RADIATION

ELEMENTARY NUCLEAR PHYSICS

1. INTRODUCTION

The modern epoch in physics may be said to have begun with the discovery of X-rays by Roentgen in 1895. This was followed by the discovery of radioactivity by Becquerel in 1896. These two discoveries led to other development in the understanding of nuclear structure, reactions, and the various types of particles involved. All these investigations finally culminated in the discovery of "Fission" which resulted in the development of atomic energy, both in peace and war. We shall review here briefly our knowledge of nuclear radiation and atomic structure.

2. ATOMIC STRUCTURE

All matter is made up of elements. The smallest part of an element is the atom. The size of a hydrogen atom, which is the smallest, has a weight of about 1.67×10^{-24} grams. In comparison, a uranium atom would weigh about 4.0×10^{-22} grams. The atom itself consists of a central nucleus surrounded by a cloud of electrons ranging in number, from one for hydrogen to ninety-two for uranium. These electrons are said to move in orbits around the central nucleus. The size of the nucleus is considerably smaller than that of the atom, the size of the hydrogen nucleus being 1.4×10^{-13} cm. The nucleus itself consists of protons which carry a unit positive charge 'e' ($e = 1.6 \times 10^{-17}$ coulombs) and neutrons which carry no charge. The mass of the neutron is only slightly greater than that of the proton. The electron carries a unit negative charge and has a mass approximately 1/1840 of that of a proton or a neutron. Hence most of the mass of the atom is carried in the nucleus. It is also known that most of the energy of the atom is also stored in the nucleus.

3. ELEMENTS AND ISOTOPES

All the elements of the periodic table are made up of protons and neutrons in the nucleus and electrons orbiting around it. The total charge carried by the nucleus is equal to the number of protons in it. This number is called the 'atomic number' and is characteristic of each element. Since the atom as a whole is electrically neutral, there will be as many electrons around the nucleus as there are protons in it. The number of protons plus neutrons gives the 'mass number' of the atom. Thus the mass number of carbon, ${}_6C^{12}$, is 12 and the atomic number is 6. If two nuclei have the same number of protons in the nucleus, but different number of neutrons, they are called isotopes. For example, ${}_4Be^7$ and ${}_4Be^{10}$ are isotopes of beryllium having the same number of protons and the number of neutrons being 3 and 6 respectively. Isotopes have the same chemical properties which depend on the number of electrons in the orbits around the nucleus, but different physical properties which depend on the central nucleus.

4. RADIOACTIVITY

As mentioned before, it was noted early in this century that certain substances are radioactive, that is, they spontaneously emit different types of radiations and transform into other elements depending on the emitted radiations. This process, which was first noted in the heavy elements like uranium, radium etc., was found to obey exponential decay law, that is, the amount of radiation emitted decreases with time in such a manner that the period for reduction to half the initial value is constant. This period is called the "half life" and is characteristic of the concerned isotope. The various types of radiations emitted in radioactive decay are described in subsequent sections.

5. ALPHA RADIATION

Alpha radiation is emitted generally by the heavy elements like uranium, thorium, radium etc. They are identical with the nucleus of the helium atom, carrying two protons and two neutrons. Hence their mass is about four times that of the nucleus of the hydrogen atom (one proton only) and the charge is +2e, that is, twice that of the hydrogen nucleus. The alpha radiation is usually highly energetic, having energies in the range of 4 to 9 Million electron volts. (1 MeV = 1.602×10^{-6} ergs). In view of its heavy mass and charge, alpha particles are highly ionizing and are easily absorbed. Hence they can travel only a few centimeters or so in air.

They cannot penetrate beyond the skin and are harmful only when they enter the body organs either by inhalation or by ingestion of contaminated food.

All the alpha radiations emitted from a nucleus will have the same energy and in some cases, two or three discrete energy values. The alpha radiations have a fixed penetration range in the various materials which is characteristic of the material and alpha energy.

Beyond this range, the alpha radiation cannot penetrate. On alpha emission, the element will transform into another element of atomic number two less and the mass number four less than those of the original element.

6. BETA RADIATION

Beta particles are particles identical to electrons. They are emitted in radioactive decay with various energies ranging from nearly zero to a maximum energy characteristic of the decaying radioactive isotope. Hence their energy spectrum differs from that of alpha rays which have discrete energies. The beta rays can travel a few feet in air but cannot penetrate much beyond the depth of the skin of a person. Hence, like the alpha rays, they are harmful only when they are inside the body. From outside they can cause only skin burns. Beta rays are absorbed in an approximately exponential manner, that is, they have a characteristic thickness in the particular material which will reduce the radiation intensity to half the initial value. This value depends on the atomic number of the material, its density and the maximum energy of the beta particles.

Although theoretically the intensity of the beta radiation cannot be reduced to zero due to the exponential absorption, it can be reduced to negligibly small values by using several "half value thicknesses" of the shielding material.

In emitting beta rays, the radioactive material transforms into another element of atomic number one higher than the original element as the nucleus loses one negative charge (or gains one positive charge). The mass number, however, remains the same as the number of nucleons in the nucleus remains the same. Some nuclei emit a positively charged beta particle, "positron". In such cases, the element transforms into an element with atomic number one lower than the original one.

7. GAMMA RADIATION

Gamma radiation is electromagnetic radiation similar to light but of much higher energy. The wavelength of gamma rays is much shorter than that of visible light. The energy of the electromagnetic radiation or photon is given as $W = h\gamma$, where h is the Planck's constant, γ (nu) is the frequency, equal to $c\lambda^{-1}$, c being the velocity of light and λ (lambda) the wave length of the radiation. Gamma rays are emitted in radioactive decay along with alpha or beta radiations. Like alpha rays, gamma rays have discrete energies. They are absorbed exponentially in materials, but in view of their great penetrating power,only thick blocks of concrete, lead or other high density materials can reduce their intensity to small value. The gamma rays can deliver a whole body dose either from outside or inside the body due to their high penetrating power.

The element does not change due to gamma radiation, but will change depending on the accompanying alpha or beta radiation.

8. NUCLEAR REACTIONS

In addition to radioactivity, the nucleus of the atom can undergo changes and emit radiations as a result of various types of "nuclear reactions". In these reactions, the nucleus is bombarded by particles like alpha rays, protons, neutrons etc., which are generated (and accelerated) in accelerators, reactors etc. The nucleus on bombardment, emits other particles like protons, neutrons, deuterons and changes accordingly. The emitted particles like protons, deuterons etc., carrying charge have fixed ranges in materials. Neutrons like the gamma rays, are exponentially absorbed and are hazardous in view of their high penetrating power. In many of the nuclear reactions, the energy put in for the reaction is more than the energy got out. That is, they are 'endoergic' and not 'exoergic'.

9. NUCLEAR FISSION

One nuclear reaction of great interest is the nuclear fission which takes place in heavy nuclei like uranium, plutonium etc. When a neutron enters the nucleus of an atom of one of the above elements, the atom splits into two nearly equal parts releasing two or more neutrons and about 200 MeV of energy. These neutrons can strike other nuclei and cause further fissions and thus a chain reaction can be built up under certain conditions.

Although many isotopes of heavy elements can fission, only a few like U-235, Pu-239, U-233 can sustain a fission chain reaction from which energy can be usefully tapped. (This is because of the possibility of fission in these isotopes from the neutrons released from their own fission. Other isotopes would require neutrons of higher energies than that of the neutrons produced in their own fission.)

If the fission reaction is controlled to maintain a steady level of fissions, we have the nuclear reactors from which energy can be usefully tapped. In nuclear weapons, however, the chain reaction is allowed to build up at a very rapid rate to create an explosion.

In the fission reaction, the uranium atom splits into two or more parts and the products formed are elements of much smaller atomic number. These fission products are highly radioactive, emitting beta and gamma rays and are very hazardous. They have half lives ranging to several years, and, depending on their chemical properties, they can enter the human body. Thus these fission products can give internal as well as external dose.

10. NUCLEAR FUSION

In nuclear fusion, light nuclei like those of the isotopes of hydrogen, fuse together to form bigger nuclei. These reactions, like fission, are exoergic and release enormous amounts of energy. A fusion process can be realized by bombarding light element by cyclotrons or other high energy accelerators, In these cases, only an extremely small fraction of the bombarding particles is utilized for fusion and hence the energy obtained is extremely small compared to the energy required for accelerating the bombarding particles. However, at extremely high temperatures, it is possible to get significant amounts of energy. The best illustration of the thermonuclear process is the formation of helium from hydrogen in the stars. Another example is the hydrogen bomb, in which using the high temperatures of the nuclear fission reaction as the trigger, a fusion chain reaction is built up to release enormous amounts of energy. However, controlled release of energy by fusion, as in fission reactors, has not yet been achieved.

A. NATURAL RADIOACTIVITY

INTRODUCTION

Man has always been exposed to low-level ionizing radiation which is present in his environment. This radiation, referred to as "natural background", is not directly caused or produced by man, but due rather to natural causes or sources.

Because the exposure which man has received from background radiation causes a few discernible effects, a knowledge of the sources of this radiation and the dose which man receives from it would be helpful in establishing, for man, levels of exposure which are not expected to have significantly harmful effects.

The sources of natural background can be separated into the two following categories:

a) Those which are found in their original or natural environment.

b) Those which are found not in their natural environment but rather have been rearranged or moved by man to an artificial environment.

a) NATURAL SOURCES OF EXPOSURE

Sources in their Natural Environment

1. Cosmic radiation

Primary cosmic rays consist of high energy protons, helium nuclei, and heavy ions, which originate outside of the earth's atmosphere, and are present throughout the universe. A small fraction of them strikes the earth's atmosphere and produces secondary radiations which give a continuous radiation dose to the earth's inhabitants. These "secondaries" consist of photons, electrons, mesons and neutrons.

The radiation intensity varies with latitude. It is about 10% lower at the geomagnetic equator than at higher latitudes of 45° or greater. Of more importance, however, it the variation of intensity with altitude. The dose at 40,000 ft. is about 100 times as great as it is at sea level. Cosmic radiation constitutes approximately 25% of the total background dosage to the population.

2. Terrestrial radiation

Several of the elements in the earth's crust are radioactive. Those which contribute most to natural background are C^{14}, K^{40}, and members of the U^{238} and Th^{232} series. On the average, one square mile of soil one foot deep contains about one ton of K^{40}, three tons of U^{238}, and six tons of Th^{232}. Surface rock contains about twice as much uranium and thorium as does the soil. Uranium is found in chemical combination in deposits of rocks and minerals in Colorado, New Mexico, Utah and other Western states; also to some extent in Tennessee and Florida. Thorium is found in large mineral bearing deposits in the Rocky Mountain States and the Carolinas. Potassium-40 is found primarily in granite and alum shales with lesser amounts in soil and sandstone.

Water and air also contain C^{14}, K^{40}, Th^{232} and U^{238} and their daughter products, though to a much lesser extent. The levels of radioactivity in the air vary considerably with location, weather, and atmospheric conditions.

b) SOURCES IN AN ARTIFICIAL ENVIRONMENT

Man disturbs some natural sources of radioactivity by removing them from their natural environment and transporting them to an artificial one. By so doing, he tends to concentrate them in some places and to diffuse them in other places.

1. Uranium mining and milling operations

Because radioactive wastes are discharged into the air and water from mining operations, these operations are considered to add to the background exposure of the population. Here, as in terrestrial radiation, the food, water, and air which man consumes contain radiocontaminants. Though there is an external exposure hazard to the mining and milling workers, the dose which the general population receives is primarily an internal one.

2. Building materials

Many of the materials which man uses for his buildings contain U^{238} and Th^{232} from the earth's crust. The uranium and thorium constitute external sources of exposure while their daughter products, beginning with the gases radon and thoron respectively, provide internal sources of exposure through the air.

The intensity of radiation which man receives from building materials depends upon the type of structure in which he dwells. The dose rates from wood, brick, and concrete are in ratios of about 1:2:3.

c) OVERALL DOSES FROM NATURAL BACKGROUND

Assuming that a person spends two-thirds of his time indoors and that the distribution of buildings in which he spends his time is divided equally, the gonadal dose rate which he would receive from external exposure ranges from about 0.55-1.05 mGy/year. This figure includes the dose rate from cosmic and terrestrial radiation as well as that from building materials. The total internal gonadal dose rate from mining and milling, terrestrial, and building material sources is about 0.23 mGy/year.

A breakdown of the doses received from the various natural background sources is as follows:

SOURCE OF RADIATION DOSE TO GONADS (mGy/year)

External	
Cosmic Rays at Sea Level	0.30-0.70
Terrestrial plus Buildings	0.25-0.75
Radon and Thoron in Air	0.02
Internal	
Potassium - K^{40}	0.20
Carbon - C^{14}	0.01
Radon, Thoron and Daughter Products	0.02
Total Background Dose	0.80-1.70 mGy/year

It should be remembered that the dose rates as listed above are only average figures giving typical dose rates under "normal" circumstances. The background dose to any person or section of the country can vary from these figures by a factor of 2 or 3.

B. ARTIFICIAL RADIOACTIVITY

NUCLEAR DETONATIONS AND FALLOUT

INTRODUCTION

The testing of various nuclear and thermonuclear devices, whether dropped from an airplane, in towers, barges, or slightly below the earth's surface, thrusts radioactive fission products and earth and water mixtures into the stratosphere (the latter occurs principally as a result of larger explosions). These particulate mixtures also include unfissioned and unfused device materials, casing and structural test materials (which may be activated by neutrons from the detonation), and remains of plant and animal life (which also may undergo neutron activation).

Depending upon the altitude and physical-chemical nature of these particles, they fall back to earth, under the influence of gravity and meteorological conditions, during a span of seconds to decades. When they make contact with the terrestrial and aquatic environment, these materials are more properly call fallout. The terms rainout and snowout may also be used as sub-categories of "fallout".

Adsorptive and absorptive transfers of fall-out into the biosphere are possible causes for concern by mankind. The ultimate deposition of fallout radionuclides in various human "critical organs" may tend to increase the incidence of various neoplasms, shorten lifespan, and accelerate normal aging processes.

a) NUCLEAR AND THERMONUCLEAR DEVICES WHICH CAN CAUSE FALLOUT

Three basic types of devices have been tested for warfare applications as bombs, rocket warheads, and artillery shells. Some of these may be modified for usage in clearing waterway channels, producing heat and radionuclides in underground mines, and other possible peace time applications.

1. Fission Device (also called atomic, or nuclear weapon)

This device utilizes only fissionable material such as uranium-235 or plutonium-239. The first one was detonated on a tower at Trinity Site, New Mexico on July 16, 1945. A blast equivalence of about 20 kilotons (KT) of trinitrotoluene (TNT) is common for such a weapon. Both gun-barrel and spherical assemblies have proven successful. The devices may be made small enough to fit into an artillery or bazooka shell.

2. Fission-Fusion Device (also called hydrogen or thermonuclear weapon)

This weapon is basically a fission device which is surrounded by the heavy isotopes of hydrogen: deuterium and tritium. The fission core releases tremendous quantities of heat and neutrons which initiate fusion reactions in the hydrogenous shell. Fusion events in turn release enormous quantities of energy and very high speed neutrons. The first hydrogen bomb was detonated by the United States in the spring of 1952.

3. Fission-Fusion-Fission Device (also called superweapon)

This device consists of a hydrogen bomb surrounded by a casing of relatively inexpensive natural uranium. Fast neutrons from the fusion are capable of fissioning the uranium, and a nominal weapon of this design may release the equivalent of 20-40 megatons(MT) of TNT. The first super bomb was detonated by the U.S. on March 1, 1954, in the Pacific Proving Grounds. Difficulties of delivery constitute essentially the only limitation upon the possible size of these weapons.

b) RADIOACTIVITY PRODUCED AND DISPERSED BY NUCLEAR DETONATIONS

Three types of radioactive material are produced and dispersed by nuclear detonations. These are: unfissioned weapon materials, mixed fission products, and materials with induced activity.

1. Unfissioned Materials

For the earlier designs, detonation efficiencies were less than 1%. This means that quantities of unfissioned uranium and plutonium may be detected near the site of detonation. The extremely toxic radio-and chemical properties of these materials indicates that their ingestion and inhalation should be avoided. In general, however, unfissioned materials will not appear in significant amounts beyond the distance at which blast and radiation effects are severe.

2. Mixed Fission Products

Millions of curies of mixed fission products are produced for each kiloton of TNT blast equivalence of a nuclear device. The primary fission fragments consist of about 60 various radionuclides and these eventually decay to form several hundred secondary fission products. This mixture of fission products, as produced by a 20 kiloton weapon, decays at a progressively slower rate with time as indicated in Table I.

Table I[27] - Total Gamma Activity of Fission Products From a 20-Kiloton Device

Time After Detonations	Activity (Megacuries)
1 minute	820,000
1 hour	6,000
1 day	133
1 week	13
1 month	2.6
1 year	0.11
10 years	0.008
100 years	0.0006

One fission product of biological importance is strontium-90. It has been estimated that all detonations by the United States, the United Kingdom, and U.S.S.R. until the latter part of 1958 produced approximately 7,000,000 curies of this radionuclide.[28]

3) Induced Radioactivity

The species and quantities of radionuclides formed by neutron bombardment depend largely upon the environment of the detonation site. Both short-and long-lived materials are formed, primarily by (n, γ) and (n, p) reactions, and their decay pattern is similar to that for fission products. Typical radionuclides formed in this manner during nuclear detonations are carbon-14, sodium-24, silicon-31, aluminum-28, chlorine-38, and cobalt-60, zinc-65, manganese-54, and scandium-46.

It has been estimated that, for air bursts, 20,000 curies of carbon-14 are produced per megaton of weapon yield. Surface detonations give only about one-half this amount of carbon-14 since not as much atmospheric nitrogen is available for the reaction. The production reaction is as follows:(2)

$$_7N^{14} + {}_0n^1 \rightarrow {}_6C^{14} + {}_1H^1$$

Excluding the 1960 nuclear bomb tests of France, detonations to date have produced about 3,000,000 curies of carbon-14 which are now distributed in the stratosphere, troposphere, ocean, and terrestrial biosphere.

It has been estimated that there are 170 megacuries of cosmic ray-produced carbon-14 in the above mentioned reservoirs before bomb testing began. Total C^{14} global activity is probably slightly in excess of 174 megacuries.

During the 1958 U.S. Pacific bomb tests, tungsten was intentionally incorporated into detonation devices so that radioactive tungsten-185 would be produced by an (n, γ) reaction. This radionuclide has provided valuable information on the rate of fallout and has been a tool for studying weather phenomena.(3)

c) DETONATION FIREBALL AND THE PHYSICAL NATURE OF FALLOUT

After a delay of a few millionths of a second from the time of initial weapon detonation, a luminous gaseous sphere, called a fireball, is formed in the extremely hot atmosphere. Depending upon weapon design and size, the maximum size of the fireball may vary from 900 feet to over three miles in diameter. Luminosity of the fireball usually does not vary greatly with the energy equivalence of the bomb.

Vaporized bomb materials, water droplets, and debris condense to form the "atomic cloud" as the fireball increases in size and cools. This cloud is red or reddish brown when first formed because of the heat catalyzed production of nitrogen dioxide, nitrous acid, and other nitrogen compounds from atmospheric gases and water vapor. Eventually the cloud changes to white due to the water droplets present in it. In surface or near surface, strong updraft called "afterwinds", sucks dirt and debris up from the ground into the radioactive cloud. As the upward thrust of debris slowly subsides, gravity begins to retrieve particulates to the earth's surface. This causes a lengthening and widening of the columnar cloud, and eventually the debris disperses widely due to the jet stream, meteorological conditions, and the continued influence of gravity.

1. Types of Fallout Particles

Fallout particles may be grouped as follows:

a) Plated-type fallout

Vaporized bomb products and induced activities which condense out as a thin shell coating on elevated particles of earth and debris form this type of fallout. Much fallout is of this nature especially if the detonation takes place at a site relatively devoid of silicate sand.

b) Fused-type fallout

Bomb materials may undergo a thermal fusion with silicate to form minute melted, glassy "beads" which contain air bubbles and mineral grains. These may be transparent to opaque and pale green or yellow to brown or black in color. Radioactivity is distributed irregularly throughout these spherical or irregular forms.

c) Droplet-type fallout

If a device is detonated near a body of water, large quantities of water may be vaporized in the fireball. As the vapor condenses out of the cooling atomic cloud, fission products and earth debris may be suspended in water droplets and then fall back to earth as precipitation.

2. Categories of Fallout

Fallout from near surface bursts may also be categorized according to the elapsed time between the time of detonation and complete descent to earth.

a) First fallout - also called Local fallout

About 80% of the radioactive particulates produced by a surface detonation is deposited within a few hours at a distance no further than a few hundred miles from the burst point. Particles which have a diameter of 340 microns, 250 microns, and 150 microns may fall from 80,000 feet in about 45 minutes, 90 minutes and 4 hours respectively.(4) It is the first fallout which constitutes the acute radiological hazard. Detonation of a single superweapon may contaminate an area of 5,000 square miles or greater to lethal levels for external beta and gamma exposures.

b) Second fallout - also called Tropospheric fallout

Fallout particles of smaller sizes and masses are deposited in the lower atmospheric layers, and are gradually brought down in a matter of one to two months by ordinary weather phenomena.

These tropospheric particulates may travel completely around the world in a shell-like fashion but they usually stay in a rather narrow radioactive band which has the general latitude of the detonation site.

c) Delayed fallout - also called Stratospheric fallout

This type of fallout is created by surface detonations of weapons of megaton size or greater which propel radioactive particulates of sub-micron diameters to heights of 16 miles or more. The estimated mean residence time of stratospheric fallout is now estimated to be 1 to 3 years. 1958 Russian bomb tests in the Arctic resulted in a stratospheric residence time of about one year.

d) DISTRIBUTION OF STRATOSPHERIC FALLOUT

Theories as to the mechanism of stratospheric fallout have been established by Dr. Willard Libby and Dr. Lester Machta. The Libby theory proposed that there is a rapid stratospheric mixing with uniform leakage back into the troposphere where fallout is deposited according to meteorological patterns. Meanwhile, Machta's fallout model has produced slow stratospheric mixing with unequal leakage back from the stratosphere as a result of tropospheric-stratospheric air mixing in the vicinity of the jet streams. Available data on surface deposition levels indicate that the Machta preferential stratospheric fallout theory is the more satisfactory explanation.

C. RADIOACTIVITY IN THE ENVIRONMENT

a) Metabolism of radiation in Man and Animals

INTRODUCTION

Many radionuclides are produced by man's use of nuclear energy. Of these, only a limited number are important as sources of internal radiation to the human body. These specific radiocontaminants can reach man by way of the terrestrial food chain. This discussion has a two-fold purpose; namely 1) to describe the characteristics that determine the environmental significance of a radionuclide and 2) to present a detailed description of the food chain behavior of several of these radionuclides of environmental significance.

CHARACTERISTICS OF RADIONUCLIDES OF ENVIRONMENTAL IMPORTANCE

In order for a radionuclide to be a significant environmental contaminant it must possess certain characteristics. These characteristics must be such that they allow the nuclide to move from its point of origin, through the food chain, and still remain a possible health hazard to man. In general the radionuclides of environmental significance are those which are readily taken up by plants and animals that are used by man. They are either isotopes of elements important in metabolism or closely similar to them. For example, the alkali metal, cesium 137 is metabolically similar to potassium. It is readily absorbed and circulates freely throughout the body irradiating all tissues. The fission product iodine 131, an isotope of the essential element iodine, concentrates in the thyroid gland. Strontium 90 and radium 226 are alkaline earths like calcium and follow it to the bone. Carbon 14 and hydrogen 3, isotopes of two very essential elements, are distributed throughout all living tissues. Because the metabolic processes of all plants and animals are similar, radionuclides which concentrate in animal tissues are usually those which pass most readily through the food chain.

A. Specific Characteristics

The relative importance of individual radionuclides depends on many factors. Among the most important factors, the following can be cited:

1. Fission yield and half-life

Radionuclides with relatively high fission-yields and moderate to long half-lives will be the most important in food chain transference. Since reduction or biological discrimination generally occurs at each step in the food chain, the radionuclides produced in the greatest abundances will have the best opportunity to reach the human population in the greatest quantity. Of necessity, these radionuclides will have to have a sufficiently long half-life to survive through the food chain in significant amounts.

2. Rate of entry and survival through the food chain

Only those radionuclides which enter the food chain at a significant rate and quantity will be of importance to man. These radionuclides must also possess characteristics that allow for their continued movement through the chain. For example, significant quantities of cesium 137 are rendered unavailable to plants in many soils due to entrapment in the lattice structure of some clays.

3. Uptake-gastrointestinal absorption by man and animals

Radionuclides which undergo significant gastrointestinal absorption by man and animals are those of greatest concern in food chain transference. These radiocontaminants are also readily transferred to animal products such as milk and meat which are used as food by man.

4. Amount deposited and retained in the critical organ

The most hazardous irradiation situation results from radionuclides that are concentrated by essential organs of the body in relatively large quantities and are retained in these organs for long periods of time. The deposition of strontium 90, a bone-seeker of long biological half-live, is an example of such a situation.

5. Type and energy of radiation emitted

The magnitude of the hazard from radionuclide deposition in the body is dependent on the type and energy of radiation. For the deposition of the same activity of a radionuclide in a given organ the hazard from the type of radiation would be alpha > beta > gamma. The hazard from the given emitter would also increase with increasing energy of emission.

B. Characteristics Based on Physical, Chemical, and Biological Properties

An arbitrary separation of the above listed characteristics into physical and biochemical categories can be made for discussion purposes.

1. Physical Properties

The fission process gives rise to a mixture of radionuclides with a wide range of half-lives. Each of these nuclides are produced in a certain proportion (abundance) which is dependent on the fissioning material and the energy of the fissioning neutrons. The abundances of the various nuclides produced have been found to be approximately the same for the different fissionable materials. Table 1 presents the most important fission products, based on fission abundance and half-life, which are of immediate concern in environmental contamination.

Table 1.[1,2] FISSION PRODUCTS IMPORTANT DUE TO FISSION ABUNDANCE AND HALF-LIFE*

(Atom Yield > 0.03 per cent; half-life > 10 hrs)

Chemical character	Isotope	Fission abundance (%)	Half-life
Halogens	^{131}I, ^{133}I, ^{135}I	3.1, 6.3, 6.0	8.1d, 22h, 6.7h
Oxygenated anions	^{132}Te-^{132}I	4.0	78h, 2.4h
Alkali metals	^{132}Cs-^{137}Ba	6.2	37y, 2.6m
Alkaline earths	^{89}Sr, ^{90}Sr-^{90}Y ^{140}Ba-^{140}La	4.6, 5.1	51d, 26y, 61h 12.8d, 40h
Rare earths	^{91}Y, ^{95}Zr-^{95}Nb ^{141}Ce, ^{144}Ce-^{144}Pr ^{143}Pr, ^{147}Nd, ^{147}Pm	-**, 6.3 6.0, 5.0 -**, 2.9, 2.7	57d, 65d, 35d 33d, 290d, 17.5m 13.7d, 11.6d, 3.7y
Noble metals	^{103}Rh, ^{106}Ru-^{106}Rh	3.4, 0.5	40d, 1.0y, 30s

*Fission abundance values are approximately the same for U^{233}, U^{235} and Pu^{239}

**Data lacking on abundance

These nuclides are characterized by a high fission abundance and a moderate too long half-life when considering parent-daughter relations. Many of these radioisotopes can subsequently be eliminated from food chain consideration because 1) they do not possess the necessary bio-chemical characteristics for efficient transfer through the food chain and 2) the half-lives are not significantly long enough to present a long-term health hazard. The more important nuclides will be those which are formed in high abundances, with a moderate to long half-lives and which are isotopes of, or chemically similar to essential elements.

2. Biochemical properties

The chemical and biological properties of the various radionuclides greatly affect their ability to move through the food chain. Table 2 presents the fission-products of biological importance grouped according to similar chemical characteristics, and shows the relative uptake by the total body and critical organ from the gastrointestinal tract.

From Table 2 it is seen that the important fission products are those which comprise the rare earths, the zirconium-niobium isotopes, the noble metals particularly ruthenium and rhodium, the isotopes of iodine, the alkali metal cesium and the alkaline earths, especially strontium and barium. Some indication of their relative importance biologically can be seen by examining the fractional uptake from the gut of these various groups of isotopes. It will be seen that although the rare earths constitute an important group of fission products, they are of little concern in the food chain due to their limited gastrointestinal absorption and concentration in the critical organ by man and animals. A similar situation exists for the isotopes of zirconium, niobium and the noble metals. This leaves the alkaline earths, strontium and barium, the alkali metal cesium and the iodine isotopes as nuclides of primary importance. Depending on the particular situation, these nuclides will assume a greater or lesser degree of importance in the food chain. Additional biological factors which aid in accessing the potential health hazard from the particular nuclide include 1) the quantity deposited and the residence time of the nuclide in the critical organ, and 2) the essentialness or indispensability of the critical organ to the organism.

Table 2.(1,2) FISSION-PRODUCTS OF BIOLOGICAL IMPORTANCE*

Chemical character	Isotopes important on account of abundance & half-life body	Gastrointestinal uptake From GI tract to:		
		Total	Critical organ	
Halogens	^{131}I, ^{133}I, ^{135}I	1.0	0.3	Thyroid
Oxygenated anions	^{132}Te-^{132}I	0.25	-	-
Alkali metals	^{132}Cs-^{137}Ba	1.0	0.4	Muscle
Alkaline earths	^{89}Sr, ^{90}Sr-^{90}Y ^{140}Ba-^{140}La	0.3 0.05	0.2 0.04	Bone Bone
Rare earths	^{91}Y, ^{95}Zr-^{95}Nb ^{141}Ce, ^{144}Ce-^{144}Pr ^{143}Pr, ^{147}Nd, ^{147}Pm	10^{-4}	$3\text{x}10^{-5}$	Bone, liver
Noble metals	^{103}Rh, ^{106}Ru-^{106}Rh	0.03	$4\text{x}10^{-3}$	Bone, liver

*Isotope pairs are classed according to the chemical and biological characteristics of the parent

SPECIFIC RADIONUCLIDES

The fission products which enter the environment from fallout or from various nuclear facilities include more than 30 radioactive nuclides. From the above discussion it is evident that all of these radionuclides are not equally harmful to the human population. Intensive study of fission product behavior in the food chain has revealed that strontium 89, strontium 90, barium 140, iodine 131 and cesium 137 are the radionuclides of major concern. Strontium 90 and cesium 137 are radionuclides of long physical half-life and are considered long term hazards. Strontium 89, barium 140 and iodine 131 due to their shorter physical half-lives are only short term hazards. This discussion will deal primarily with the environmental behavior of strontium 90, iodine 131 and cesium 137. Other radionuclides of concern (e.g., barium 140, strontium 89, radium 226, carbon 14, etc.) will only receive brief consideration.

A. Fission-Product Alkaline Earth Metals

The radionuclides of major importance in the alkaline earth series are strontium 89, strontium 90 and barium 140. Their half-life characteristics are tabulated in Table 3. Like calcium, these alkaline earth radionuclides are deposited in large amounts in the skeleton. All three radionuclides are produced in relatively large abundance during nuclear fission (see Table 1) and assume a greater or lesser degree of importance in food chain contamination depending on the time period considered after the contaminating event. Strontium 89 and strontium 90 behave the same chemically so that the food chain behavior of one applies to the other as well.

The main terrestrial pathways for radio-strontium (i.e., strontium 89 and strontium 90) are shown in Figure 1.

The relative importance of one pathway over that of another with respect to population body burdens does not only depend on the particular isotope of radiostrontium but also on dietary characteristics of the particular population such as the type and quantity of food ingested, geographical location, degree of food processing and economic status.

As an example of the behavior of a particular radionuclide in the food chain, strontium 90's movement in the pathway atmosphere to soil to plant to animal to milk to man can be cited. As a given quantity of strontium 90 moves through this pathway, the various environmental components, including man, tend to discriminate or reduce the quantity which is finally available for bone deposition in man. The strontium 90 is deposited on the soil and effectively diluted with soil constituents before being assimilated by the plant. The grazing animal received the contaminant through ingestion of various vegetative forms and reduces the quantity of strontium 90 excreted into the milk by inherent discriminatory processes. The remaining strontium 90 is then available for deposition in man.

The main terrestrial pathway for barium 140 is presented in Figure 2. Because of barium 140's short physical half-life (12.8 days) it is generally of lesser importance as an environmental contaminant. Its occurrence in environmental media indicates the presence of relatively fresh fission products.

Table 3. Characteristics of Alkaline Earth Radionuclides(3,4)

Radionuclide		**Half-lives**	
	Physical	Biological	Effective
Strontium 89	51 days	50 years	50 days
Strontium 90	28 years	50 years	18 years
Barium 140	13 days	65 days	11 days

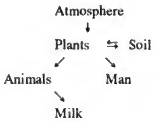

Figure 1. Main Terrestrial Pathways in the Food Chain for Radiostrontium(5)

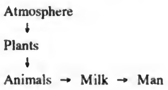

Figure 2. Main Terrestrial Pathway in the Food Chain for Barium 140(5)

Before discussing alkaline earth radionuclide behavior in the various food chain steps, a discussion of how their movement is quantitatively measured is needed in order to understand overall food chain transference. Their movement in biological systems can be considered in terms of particular radionuclide itself or in terms of comparative movement with stable calcium. The choice of measurement and mode of data expression is governed by the objective of the particular investigation. By far, the most widespread method of measuring alkaline earth radionuclide behavior is to relate it to the behavior of stable calcium. This is particularly true for radiostrontium. The method involves measuring strontium-calcium rations in the various samples (e.g., soil, milk, diet, bone, etc.) and observing the resulting changes in these ratios at various steps in the food chain. Comar, et al.[6] have defined the term "Observed Ratio" (OR) to describe the comparative behavior of strontium-calcium ratios that exist at equilibrium between the precursor and the sample (Equation 1).

$$\text{sample/precursor} = \frac{(\text{Sr/Ca})\text{sample}}{(\text{Sr/Ca})\text{precusor}} \qquad (1)$$

The observed ratio has been used to describe the overall discrimination that occurs between two environmental media or a number of physiological processes (e.g., the discrimination that occurs when the element pair passes from soil(precusor) to the plant(sample) or when the element pair passes from man's diet(precursor) to his bone(sample). It is actually a measure of the relative affinity of a environmental media or physiological process for strontium or calcium. For example, if the strontium-calcium ratio in one individual bone is 2 picocuries of strontium 90 per gram of calcium and the strontium-calcium ratio in the diet is 8 picocuries of strontium 90 per gram of calcium the observed ratio is 0.25 (i.e., 2 pCi per gram/8 pCi per gram). This result suggests that a discrimination exists against strontium in favor of calcium in a ratio of 1:4 in gram from diet to the bone. For many food chain transfer processes the OR has been found to be less than 1, implying that in these processes discrimination in favor of the passage of calcium occurs. Rearrangement of the observed ratio expression for the case of diet to bone yields the expression

$$= \frac{\%\ \text{Strontium 90 recention(body-diet)}}{\%\ \text{Calcium retention (body-diet)}} \qquad (2)$$

In this form the OR is a measurement of the percent strontium 90 retention divided by the percent calcium retention in going from the diet to body.

In contrast to the OR which indicates overall discrimination by one or more processes, the Discrimination Factor (DF) describes the discrimination produced by one physiological process. For example, $DF_{\text{absorptive}}$ designates the contribution of overall discrimination in the body that results from differential handling of the two elements by the absorptive process. Formulae have been developed to determine the DF's for the most important physiological discriminatory processes (gastrointestinal absorption, renal excretion, placental transfer and mammary secretion) in a fashion similar to those for observed ratios.

The relationship between the OR and DF is

$$\text{OR} = (\text{DF}_1)\ (\text{DF}_2)\ (\text{DF}_3)\ldots.(\text{DF}_n) \qquad (3)$$

Thus the OR is the product of the total number of discrimination factors operative. When a single physiological process is involved the OR and DF are identical.

The validity and use of observed ratios in describing radiostrontium behavior in the food chain depends on many factors. Several of these deserve brief mention at this time for an initial understanding of the problem. At the domestic animal and human level of the food chain, it is well documented that calcium metabolism is under homeostatic control.[6,7] This means that variations in dietary calcium within normal physiological limits does not alter the constance of the calcium levels in blood, milk and the total body. In contrast to calcium behavior, strontium metabolism is not under homeostatic control (not regulated by the normal amounts of stable strontium) but rather appears to be regulated by the calcium level or perhaps the total alkaline earth level. Thus under normal dietary conditions the calcium level of the various body fluids and the total body content are relatively constant whereas the stable strontium level appears to be regulated by one or more of the dietary levels of strontium, calcium or total alkaline earth. Whether just one of these factors or all three are responsible is not known. An important generalization from the above is that within normal dietary ranges the stable strontium to calcium ratio and the strontium 90 to calcium ratio in the body tissues or fluids will be directly related to the ratios that exists in the diet. This had led to the development of quantitative expressions such as the OR and DF for these ratios.

The practical application of OR values depend upon their constancy. It has been shown that the discrimination occurring between food chain components can change as an interrelated function of age, diet, and time.(8,9,10,11,12) However, the OR values appear to be reasonably constant under normal dietary conditions and are consistent enough to be used for prediction purposes. This is especially helpful in the case of predicting strontium 90 bone deposition in the population since widespread dietary and skeletal burden information is limited. This is probably the greatest advantage of the OR concept - the ability to predict radiostrontium movement and concentration in food chain media that are difficult and/or economically infeasible to quantitatively assay. The use of OR is also facilitated by the fact that many aspects of calcium metabolism are well known. Of course the assumptions and limitations inherent in the use of the OR concept for prediction purposes should always be kept in mind. A few examples will illustrate the prediction value of this concept.

The OR concept may be used to integrate samples for an estimate of total Sr^{90}/Ca ratio intake. The Sr^{90}/Ca ratio intake of the dairy cow can be estimated by calculation from milk values. By dividing the Sr^{90}/Ca ratio in milk by 0.1 (a typical value for OR milk, diet for the dairy cow) would give an estimate of the Sr^{90}/Ca ratio of the cow's diet ($Sr/Ca_{(diet)} = Sr/Ca_{(milk)}/OR$). This estimation of dietary intake is surely simpler and in many cases would be more accurate than measuring total intake directly or summing up the strontium 90 and calcium contributions from feed and water. The use of the OR concept in this case assumes a constant OR value for a diet which is considered typical. These assumptions appear quite well founded.(6) On the other hand, the prediction assumes that the cow's calcium intake is uniformly labeled with strontium 90, that the animal is in equilibrium with his diet and that the strontium 90 and calcium in the milk comes primarily from that of the diet. These conditions may or may not be operative depending on the particular circumstances.(1,13,14)

A second use of the OR concept is the prediction of human bone burdens of strontium 90. The most direct approach to the problem would be to establish retention curves. Theoretically, net retention of strontium 90 in bone is expected to have a high initial value governed by the absorptive mechanism and then fall to zero as the individual approaches equilibrium conditions with the intake. Studies such as indicated in Figure 3 have shown that the net retention of stable strontium in man is zero at steady state.

It is difficult to estimate body burdens since the retention of strontium 90 is primarily a function of time of dietary intake. It is also a function of dietary history, nutrition, age, and history of past intake of strontium 90. To use this approach retention curves for varying condition of age, dietary history and nutrition would have to be determined, and if such curves were available it would then be necessary to determine integrated life-time values of retention. Thus a specific value for strontium 90 has little meaning unless it can be related to the above factors and some unit of bone. Returning now to the OR for bone burden predictions, the strontium 90 to calcium ratio in the bone can be estimated by using the $OR_{(bone/diet)}$ for the human and an estimate of the dietary strontium 90 to calcium ratio. The $OR_{(bone/diet)}$ for the human has been estimated from animal stable strontium-calcium ratios and tracer studies. Dietary surveys have been used in estimating the strontium 90 and calcium intake. Once an estimate of the strontium 90 per gram of calcium has been established for the bone, the strontium 90 content of the skeleton can be determined from the total calcium, a value which is relatively well known. Of course a prediction of this type has a number of limitations which should be borne in mind. Especially in the case where the estimated bone burdens are going to be used in subsequent dose calculations. The OR calculation here assumes a skeleton formed entirely from a diet of a given strontium 90 to calcium ratio which is in equilibrium with the bone. With reference to the adult human population and the length of time strontium 90 has been in the environment, this is not the case since strontium 90 has not been present long enough to uniformly label the adult skeleton or attain equilibrium conditions. The use of the OR calculation for the adult population then will most probably represent the maximum local concentration in areas of newly forming or exchangeable bone at equilibrium conditions. On the other hand, children who have been exposed to a relatively constant dietary strontium 90 to calcium ratio will have skeletons that are more uniformly labeled than those of adults. They would also be expected to closer to true equilibrium conditions. However, as a result of other problems such as determining children's intake, a lesser degree of discrimination in the very young, strontium 90 contribution from the mother during the fetal period, etc., predictions of children's bone burdens will perhaps only be of a little greater accuracy than those for adults.

Another isotope pair has been considered in describing strontium 90 movement in the food chain - the specific activity of strontium 90 (i.e., the ratio of strontium 90 to stable strontium). It has been shown that the strontium 90 to stable strontium ratio of the diet will be identical to the strontium 90 to stable strontium ratio of the tissue (bone) that has been formed from that diet (for man).(6) Thus the OR concept could be used for these ratios in a manner similar to that for strontium 90 and calcium. However, a strontium 90 to stable strontium value for bone is not easily related to the total amount of strontium 90 in the total skeleton because the stable strontium content of bone is variable. The stable strontium content of bone depends on the stable strontium and calcium intake - a result of the lack of strontium homeostasis. Thus, even if the strontium 90 to stable strontium ratio of the diet is known, a value of strontium 90 or stable strontium per unit of bone or calcium is required to obtain the amount retained in bone. Because data on stable strontium intake and bone content is limited, and both of these parameters can be quite variable, the strontium 90 to stable strontium ratio provides little information that is useful for prediction purposes on the amount of strontium 90 retained in bone.

There are several general conclusions that can be drawn from the foregoing discussion on measurement of radiostrontium in the food chain. The OR concept appears to be the most satisfactory method at present to predict overall radiostrontium movement in the food chain, especially on a long term behavioral basis related to bone deposition in the human population. Other methods such as strontium 90 to stable strontium ratios and absolute concentration values (e.g., pCi/l, etc.), suffer from the disadvantages of limited baseline data or the fact that they give information only on one point in time. Comparison of human body burdens predicted from the OR concept to those measured from limited dietary and associated bone analysis compare reasonable well.(15) In general the measured bone levels are somewhat lower than those predicted from the OR concept. It appears that radiostrontium behavior in the food chain should be reported both in terms of strontium-calcium ratios and in absolute values so that maximum use of the data can be obtained.

1. Behavior in soils and plants

Strontium 90 may enter the food chain through absorption by plant roots or direct contamination by aerial deposition on above ground plant structures. The contaminated plants may be consumed by man directly or indirectly through his use of animal products (Figure 1). Strontium 89 and barium 140 are only transferred up the food chain in significant quantities by initial aerial deposition on above ground plant parts (short half-lives restrict soil absorption). With respect to strontium 90, it is important to distinguish between root uptake and aerial deposition since the amount absorbed from the soil depends on the cumulative deposit, while the extent of direct contamination depends on the quantity of recent deposition. This section will deal primarily with strontium 90 behavior since this radionuclide represents the most hazardous food chain situation.

a) Strontium 90 absorption from the soil.

Field experiments indicate that, depending on the soil type, the strontium 90 to calcium ratio is likely to be in the range of 1-10 pCi of strontium 90 per gram of calcium if 1 millicurie of strontium 90 per square kilometer is present in the soil.(15) The amount taken up by crops each year ranges from 0.2 to 3 per cent of that in the soil; the exact quantity being dependent on a number of soil characteristics.(15)

The most important factor determining the extent of strontium 90 uptake by plants is the quantity of labile* calcium present in the soil. The plant uptake of strontium 90 is greatest in soils of low calcium content. Addition of lime to these soils will generally reduce the strontium 90 uptake, but usually not by more than a factor of 3. When soils contain quantities of calcium that result in optimal plant growth addition of more lime has little or no effect. Soil characteristics other than calcium content usually have comparable effects on strontium and calcium absorption; thus little difference in their ratio is realized in the plant. The addition of organic matter and fertilizers in large quantities has varying and complex effects. However, when these materials are applied in the usual agricultural quantities little effect is obtained. Although various plant species show a wide variation in their ability to absorb strontium 90, it is well documented that this characteristic is correlated with the ability to absorb calcium.

Additional factors which affect the absorption of strontium and calcium from the soil are the clay and humus content, the pH, the concentration of electrolytes other than calcium and the moisture content. In general, it is possible to predict how certain characteristics of the soil influence the uptake of strontium 90. However, it is not possible to give all the details on strontium 90 uptake from a particular soil or field without extensive knowledge of all factors which influence its behavior. For example, light sandy soils will allow strontium 90 to penetrate to a deeper extent than in heavier clay soils. However, a relatively greater part of the roots will be in contact with strontium 90 in the sandy soils due to its deeper penetration. On the other hand, the strontium will also be diluted with more calcium because of the greater depth of penetration. In this case, these two mechanisms are working in opposition. In general, light soils (sandy soils in the temperate and latosol type of soil in the tropics) are poor in calcium. The strontium-calcium ratio in these soils will thus be higher than that of heavy soils (clay soils in the temperate regions and margalites in the tropics).

Another area of strontium 90 behavior that has received considerable investigation is that of movement in the soil. Downward movement in undisturbed soils is slow and even after several years 50 per cent or more of the added strontium 90 may be in the upper 5 centimeters of the soil.(17) The rate of downward movement depends on the soil type, clay and humus content, electrolyte concentration and degree of water penetration. The exact nature of the movement is unknown but it is believed that movement with water and self diffusion

*Labile calcium = calcium present in soil solution + echangeable calcium present to replenish the soil solution may be involved. In agricultural practices, cultivation of the soil causes the strontium 90 to become more uniformly distributed in the ploughed layer. Depth of cultivation combined with the rooting characteristics of the crop (deep versus shallow) is of primary importance in determining the degree of strontium 90 intake. For example, ploughing to a depth of 20 to 30 centimeters has resulted in a reduction of strontium 90 uptake by a factor of 3 when compared to surface application for shallow rooted crops such as rye, grass or kale.(15) For more deeply rooted crops the effect is smaller.

Considerable investigation has been given to the possibility that with the passage of time strontium 90 may become fixed in the soil in a less soluble form and be less available for plant uptake. Some investigators have found no evidence of fixation while others report a small quantity of fixed strontium 90 over a period of three or more years.(15, 17) It is possible that this effect varies considerably from one soil to the next. However, present evidence indicates that fixation of strontium 90 by soils is unlikely to reduce its availability to plants to any significant extent over a period of several years.

Results from water culture studies indicate that the Observed Ratio (plant shoot/solution) is close to 1 (Table 4).(15)

This value is generally applicable to plants growing in soil when the strontium is uniformly distributed or when differences in root depths are compensated for. Different strontium to calcium ratios have been observed in different tissues of the plant and this indicates a differential movement of the two ions. When the two elements enter the plant through the roots, the resulting ratio in the aerial organs most distant from the root is in general lower than the average in the plant. The ratio of strontium 90 to calcium in the vegetative tissues is often about half of that in the roots and twice that in grain and seeds. Stems may have considerable higher ratios than other plant structure. The most detailed study of the differential movement of the two ions between tissues has been that of whole grain. The ratio in wheat flour (seed portion of grain) has been reported to be 0.7 of that in the whole grain.(15) These plant discriminatory processes have little effect on the strontium 90 to calcium ratio in man's diet. Most human dietary calcium in the Western Hemisphere comes directly or indirectly from milk via leaf tissue which has a strontium 90 to calcium ratio similar to that of the rooting medium.

Different values have been reported for the OR (plant shoot/soil), but this variation can be attributed to the difficulties in estimating the availability of strontium 90 and calcium to the plant from the different soils. Because of the non-uniform distribution and the difficulties of determining the availability of the two ions in soil that are actually available for plant absorption, the strontium 90 to calcium ratio in the plant prediction from the OR concept is probably not a true reflection of what is available from the soil. Also, the lack of steady state conditions in the soil will normally limit the use of the $OR_{(plant/soil)}$ for prediction purposes on a wide basis.

b) Direct contamination of plants by strontium 90

In contrast to soil absorption where only strontium 90 is of significance, all three important alkaline earth radionuclides, strontium 89, strontium 90 and barium 140, may be transferred to the human population through direct deposition on vegetative forms. Direct contamination can be divided into two categories: 1) superficial contamination in which the radioactive material adheres to the crop surface, and 2) direct inside contamination which results from absorption through the leaves, flowers and stems with subsequent movement to other plant parts. Superficial contamination is of major importance where food and animal feed are consumed fresh. In the case of human consumption a relatively large portion of this contamination can be washed off, however, domestic animals will be presented with the total deposit for food chain transference. Direct inside contamination is dependent on the transport characteristics of the alkaline earth elements. For several of the elements, little is known about these characteristics. For calcium it is known that the transpiration* stream is the prime mover. Strontium 89 and strontium 90 are expected to behave in a similar manner. This theory has been confirmed by field and laboratory experiments in that the strontium 90 that enters through the roots is distributed to the aerial plant parts by the transpiration stream, but that which is deposited on the aerial structures is only slightly distributed to other tissues. In direct contamination the calcium content of the plant is another major factor in affecting the resultant Sr/Ca ratio since the entering aerial strontium 90 does not have the benefit of dilution with soil constituents. The plant calcium content, is somewhat dependent on the plant species. In general, the calcium content of the monocotyledons (plants containing one seed leaf in embryonic stage) is lower than that of dicotyledons (plants containing two seed leaves in embryonic stage). Most of our vegetables are dicotyledons of relatively high calcium content. Clover and weeds (animal feed) are dicotyledons while the real grasses are monocotyledons.

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*One of the primary mechanisms causing the rise of water in plants (loss of water from plants)

Table 4. Strontium-Calcium Observed Ratios in Absorption by Plants[15]

Species	Method	$OR_{(plant\ shoot/solution)}$
Barley	^{89}Sr, ^{45}Ca	~1
Ten Species	^{89}Sr, ^{45}Ca	0.7-1.3
Tomato	Stable Sr, Ca	~1
Five Species	^{89}Sr, Stable Ca	1.1
Wheat	^{90}Sr, Stable Ca	1.0
Pea	In sand culture	0.9

Direct contamination has also been subdivided according to the organs of the plant contaminated. Thus, there is floral, foliar and plant-base contamination by radiostrontium. Floral deposition is of major importance with crops such as wheat and grain which are able to entrap considerable quantities of radiostrontium (also cesium 137) in their inflorescences. This entrapment results in a higher contamination of the grain than other plant structures. Other crops such as corn and beans do not exhibit this characteristic because their flowering parts are covered with sheaths which prevent floral contamination.

Foliar deposition is most significant for vegetables and grasses. The amount of radiostrontium retained on vegetation by this mechanism depends on the extent and duration of rainfall, and the extend and structure of the aerial plant parts. Experimental studies indicate that about one-quarter of the deposit that comes down in rain may initially lodge on the vegetation of permanent pastures consumed by cattle.[15] Leaf absorption is slow and the material is readily lost in rain. Plant-base contamination is significant in the case of perennial pastures which are able to absorb radiostrontium essentially all year long. Radiostrontium on stems and leaves can be washed down to the base of plants where it will be taken up by stems and surface roots without benefit of soil dilution. Radiostrontium uptake in this manner results in transport to all plant parts via the transpiration stream.

2. Metabolism in domestic farm animals

Since animal products are quite important in the human diet, the role of domestic animals in the food chain must be studied in detail. Again the alkaline earth fission-products metabolism is similar to that of calcium, a mineral element of major importance in animal metabolism. Environmental surveys have indicated that this group is transferred most rapidly to man through the pathways atmosphere-plants-animals-milk-man and atmosphere-plants-man. One pathway may assume a greater or lesser degree of importance depending on the kind and quantity of food comprising the diet and the fission-product and calcium content of the foods of the diet. Numerous studies have shown that the alkaline earth radionuclides are secreted into the milk of domestic farm animals in significant quantities. Since this food item is the major component of the diet of young children (considered to be the most sensitive population group), and is consumed relatively soon after contamination, it has been the food most often analyzed for an indication of any environmental contamination. Thus, many studies have centered on the mechanisms and quantities of alkaline earth radionuclides secreted into the milk of animals that are used by the human population. Other major areas of study include the tissue distribution of this group of elements in farm animals subjected to acute and chronic ingestion, determination of metabolic discriminatory processes using the OR and DF concept, and determining the effect of various dietary modifications on alkaline earth behavior in the animal and its products.

a) Tissue distribution of the alkaline earth radionuclides

Two intake modes are of major importance for the alkaline earth radionuclides. First, a single or "one shot" intake of the radionuclides. This has application to the environmental contamination situation in which the animals are allowed to graze on a contaminated pasture for a few hours and then placed on uncontaminated feed. A typical distribution of an oral dose of several alkaline earth radionuclides is shown in Table 5.[16]

The distributions are contrasted to calcium 45 as a reference. The values of Table 5 should be considered as relative orders of magnitude involved and are somewhat dependent on both the experimental method employed and the time of experimentation. In general, the same results are obtained when the radionuclides are ingested as mixed fission-products, separate radioisotopes or incorporated into plant and grain materials with physical mixing. The data of Table 5 indicate that more radiostrontium than barium is secreted into the milk of the cow. This results from greater discrimination by the cow against barium 140 in contrast to radiostrontium. Fecal excretion is seen to be the major route of elimination for both barium 140 and radiostrontium. After a single administration of radiostrontium, the levels in milk will steadily rise until a peak occurs at about 30 hours. For the next 5 days the levels decrease with an effective half-life of 30 to 40 hours. Barium 140 behavior is similar with an effective half-life of about 45 hours.

Table 5. Distribution of ^{140}Ba, ^{89,90}Sr and ^{45}Ca in the Cow and Goat(16)

Material	Cow			Goat	
	^{140}Ba	^{89,90}Sr	^{45}Ca	^{89,90}Sr	^{45}Ca
Urine*	1.0	1.5	1.5	2.4	2.1
Feces*	98	95	78	93	77
Milk*	0.08-0.6	0.9	9.0	0.6	5.3
Milk**	0.01	0.1	0.9	1.6	9.9

*Values expressed as percent of dose summed over a 7 day period
**Values expressed as percent daily dose/liter after equilibrium

The second intake mode of interest is that of a constant intake over an extended period of time. When cows are subjected to chronic ingestion of radiostrontium the equilibrium distribution of the daily intake of the element is approximately the same as for the value for a single dose summed over a seven day period (the values of Table 5). Therefore the values in Table 5 can be considered as daily excretions after equilibrium has been reached. Milk values reach a plateau in about six days indicating that the animal is in near equilibrium with the diet. If contaminated feeding is continued with no further addition of radiostrontium and barium 140 to the feed, the radiostrontium and barium 140 levels will decrease with effective half-lives corresponding to the physical half-lives of the isotopes. The total amount of radiostrontium secreted into the milk ranges from 0.5 to 2 percent of the daily intake.(16) This is a reflection of variation in milk yields since the quantity of radiostrontium secreted per liter is relatively constant at about 0.1 percent of the daily intake.

The distribution of the radiostrontium in the goat is also shown in Table 5. The distribution is similar to the cow except that a much greater amount of calcium and radiostrontium are secreted in the milk of the goat. This is a result of the fact that the goat secretes a much larger quantity of the dietary calcium and radiostrontium into each liter of milk.

b) Determination of OR and DF Factors

Observed Ratios and Discrimination factors have been established for a number of metabolic processes in domestic animals. Experimental studies in this area have determined the major metabolic processes which are active in discrimination against alkaline earth radionuclides. Extrapolation of these data along with limited studies on the human population have yielded valuable information on the behavior of these radionuclides in man. Table 6 presents selected observed ratios and discriminatory factors that have been determined for animals and man.

The OR's and DF's of Table 6 have been calculated from the use of radioisotopes of barium, strontium, and calcium; strontium 90 and stable calcium. Discrimination factors have yielded important information on the physiological mechanisms that are important in the differential movement of the alkaline earth radionuclides. The more significant physiological processes that aid in the body's discriminating against strontium in favor of calcium are: 1) absorption from gastrointestinal tract, 2) urinary excretion, 3) transfer across the plancenta and 4) secretion into milk (Table 6). The processes that involve physiological handling and active transport across biological membranes are thought to be the most important.

Table 6. **Selected OR and DF Values(6, 18, 19)**

Material	Cow		Goat	Rat	Man
	Ba/Ca	Sr/Ca	Sr/Ca	Sr/Ca	Sr/Ca
OR(bone/diet)	--	0.20	0.20	0.28	0.25
OR(milk/diet)	0.05	0.11	0.12	--	0.10
OR(fetus/mother's diet)	--	--	--	0.17	0.13
OR(fetus/mother)	--	--	--	0.65	0.6
OR(plasma/diet)	--	0.18	0.29	--	--
OR(urine/diet)	2.8	1.6	1.5	3.0	0.6
OR(feces/diet)	1.4	1.3	1.2	--	--
DF(absorptive)	0.12	0.24	0.24	0.5	0.70
DF(urinary)	0.70	0.80	0.60	0.80	0.70
DF(Fecal)	0.76	0.98	--	--	--
DF(lactional milk)	0.70	0.64	0.61	--	--

The observed ratios of major practical importance for prediction purposes are those for (bone/diet), (milk/diet) and (fetus/mother's diet). Laboratory studies and dietary surveys of strontium 90 to calcium ratios in diets and bones have indicated that the OR (bone/diet) is close to 0.25 (ranges 0.17 - 0.54) for adult mammals (including humans) which are on normal diets.(19) This is the value used most often for prediction of bone levels and implies that there is a four-fold reduction in the strontium 90 to calcium ratio in going from diet to bone. For adults this strontium 90 to calcium ratio represents the maximum concentration at the sites of active bone formation or exchange at steady-state conditions. Calculation of OR (bone/diet) values for adults from fallout data is not possible because of the short time strontium 90 has been in the environment with respect to the period during which the bones of adults have been formed.

c) Chronic exposure over long periods

There are two practical chronic exposure situations with respect to intake of dietary radiostrontium (especially for long-lived strontium 90). The first situation applies to adults who have had a relatively short exposure time to environmentally deposited strontium 90 relative to the length of time that their bones have been formed. The second situation applies to children who have had a life-time exposure to environmentally deposited strontium 90.

For an adult exposed to dietary strontium 90, approximately 20 percent of the ingested radionuclide may initially be deposited in the skeleton. As ingestion continues the net retention decreases to zero as the skeleton becomes labeled under constant intake. Those initially exposed as adults, most probably, do not reach this steady-state condition during their lifetime. The amount of strontium 90 retained depends on the past history of the individual in relation to this dietary intake.

To explain strontium 90 behavior in children requires consideration of the growth and turnover of the skeleton with respect to calcium and strontium. These concepts are briefly summarized in the following paragraphs.(20, 21, 22)

1. The infant child is born with approximately 30 grams of calcium in the body and a strontium concentration of about 200 micrograms per gram of calcium. At this time the infant has an urgent need for phosphate for its soft tissues and if the dietary phosphate is not adequate, as in the case of breast feeding, calcium and strontium phosphates are utilized from the skelton. A constant bodily calcium level is maintained but a negative strontium balance (excretion > intake) may result.

2. The newborn's kidneys are able to discriminate between strontium and calcium; however, the intestinal mucosa's discriminatory ability may not be as great as that of the adult. Therefore, at this age the infant's discriminatory processes favoring retention of calcium and discharge of strontium are less than in later life. However, this lesser ability to discriminate between the two ions at this age is partially offset by the lower initial strontium 90 to calcium ratio of the fetus due to the discrimination of the mother's plancenta, and if the child is breast fed the strontium 90 to calcium ratio of his intake is appreciably less than that of older age groups.

3. During the growth in infancy there is a fast accretion of mineral into the skeleton, a threefold

increase in the first year of life. There is probably a complete remodeling of the bones. The exact amount of mineral reused and replaced with dietary sources is not known. Values from 30 to 100 percent have been predicted for the first and second years of life. This high turnover rate results in an equilibration of bone mineral with dietary sources and leads to a relatively uniform distribution of strontium 90 in the skeleton. The concentration of strontium and strontium 90 in terms of calcium will be approximately one half of that of the diet (at age 1).

4. As the infant grows the rate of accretion and turnover of existing bone decreases. At the same time the discrimination against strontium in favor of calcium approaches that of the adult. By age 7 the skeletal mass has increased by less than threefold since age 1. The strontium concentration in terms of calcium is about the same as in infancy and the strontium 90 concentration is less than in infancy.

5. From age 7 onward acquisition of new bone rises again but turnover appears to be minimal. Thus, at around puberty growth appears to be a matter of adding on new mineral to existing bone. At about this time the stable strontium intake of the child approaches the adult value. Also, at this time the relative distribution of absorbed calcium and strontium between bone and excreta alters markedly with these absorbed minerals being shunted more to excreta and less to bone. Up to this time, strontium 90 in bone has been related to the proportion of the total life span which has been spent in the contaminated surroundings. There has been a continued decrease of strontium 90 concentration in bone with age.

6. On attaining adult life acquisition of bone mineral ceases but a certain amount of maintenance or replacement continues. The adult healthy individual is in equilibrium with his mineral intake; that is absorption of calcium and strontium just balances excretion. In old age, and in cases of demineralizing diseases, there may be a net loss of minerals. The above concepts have been derived from planned experiments in the laboratory or from survey data acquired from monitoring strontium 90. These experiments and surveys have aided much in the understanding of the dynamics of mineral metabolism; however, the full picture is far from complete.

B. Radioiodine

In nuclear fission a number of radioisotopes of iodine are formed. Among the most prominent are iodine 131, half-life 8.1 days; iodine 132, half-life 2.2 hours; and iodine 133, half-life 21 hours. In fresh fission-products, the shorter-lived radioiodine isotopes will initially make the major exposure contribution because of their greater abundance. However, in older fission-products (on the order of a few days), the shorter-lived iodine isotopes will have decayed, and iodine 131 will be the radionuclide of major concern. Thus, the study of the environmental behavior of radioiodine has been concentrated on iodine 131 which has a relatively long half-life compared with the other iodine isotopes.

The major pathways of iodine 131 in the food chain are illustrated in Figure 3.

Atmosphere → Plants → Animals → Milk → Man

Figure 3. Main Terrestrial Pathways in the Food Chain for Iodine 131(5)

Because of the relatively short half-life of iodine 131, the soil pathway is not important. Since iodine 131 is only of major concern for relatively short periods of time following the deposition of fresh fission-products, it is only necessary to consider dietary foods which reach man shortly after contamination. Milk is the primary example of such a food. Experimental studies and dietary surveys have indicated that milk is the only food product that contributes a significant amount of iodine 131 to the human diet. Thus the most important pathway for iodine 131 is atmosphere-plants-animals-milk-man. It is possible that the pathway atmosphere-plants-man could result in ingestion of significant amounts of iodine 131 from unwashed fruits and vegetables that have been exposed to surface contamination. However, exposure by this route would most probably be only significant on a local or individual basis.

The quantity of iodine 131 reaching the population is dependent on the extent of deposition on the plants that lactating animals consume. This deposition may vary greatly due to the difference in the plants and the extent to which deposited iodine 131 is removed from the plants by weathering. Discriminatory processes of the grazing animal tend to reduce the final quantity of iodine 131 that reaches the population. The amount of discrimination by the animal may be quite variable and is a function of many factors (feeding practice, season of the year, type of intake, etc.)

1. Behavior in soils and plants

As mentioned previously, absorption of iodine 131 from the soil is for all practical purposes unimportant since it will disappear through radioactive decay before it can be taken up by plant roots and transmitted to man. Iodine 131 reaches the soil as an anion. Since anions are practically non-adsorbed by soils rapid loss of iodine 131 from soils would be predicted. However, a rather high retention (60 to 80 percent) of iodine 131 in soils has been reported by some researchers.(16) This discrepancy may be due to the difficulty of extrapolating the results obtained with high concentrations to circumstances where low concentrations occur.(16) More study is needed in this area. From the viewpoint of food chain pathways, it will usually only be necessary to consider foliar absorption on plants. The pathway of floral absorption will be of little importance because of the usual delay between the harvesting of the crop and its consumption. Since man's intake of iodine 131 is primarily from milk, the most important question from a practical standpoint is the extent to which iodine 131 is retained on vegetation consumed by cattle.

Due to many factors, among which are the manner of pasture management, physical and chemical state of iodine 131 and the different nature of the grazing animals, it is difficult to make many generalizations which are broadly applicable to this question. However, laboratory and field studies on the deposition characteristics of iodine 131 on vegetation indicate that 1) the rate of deposition of iodine 131 is quite variable as a result of differences in the physical state of the radionuclide, 2) deposited iodine 131 exhibits a chemical state that is not well understood; however, it is known that it is difficult to wash off a major fraction of the radioiodine, 3) once deposited iodine 131 will be readily transported to other plant structures, and 4) during precipitation iodine 131 will be deposited at a much faster rate than in dry weather, but rain also washes the plant and most probably removes some unknown amount of the radionuclide (there have been no systematic studies in this area).(23) Care should be exercised in applying information gained in investigations at one atomic installation of that of another. For example, it is not known how applicable findings at the Hanford Atomic Works, a semi-arid region, would be to a grassland area of the Midwest or Northeast.

2. Metabolism in domestic farm animals

Since milk is the major source of iodine 131 to the population, the secretion of this radionuclide into the milk of lactating animals has received major study. Other areas that have received considerable attention are the distribution of iodine 131 in animal tissues and secretion, and the factors that modify iodine 131 metabolism.

a) Secretion of iodine 131 into milk

Two experimental approaches have commonly been used to study the secretion of radioiodine into milk. A single oral administration of radioiodine has been used to simulate the environmental situation of short exposure to the grazing animal such as might occur after a brief escape of radioiodine from a reactor. The second approach consists of orally administrating radioiodine over long periods of time to simulate the situation where the feed is contaminated, and it is impossible or impractical to provide uncontaminated feed. The laboratory approach is believed not to differ significantly from that where radioiodine is contained in fallout.

b) Distribution of iodine 131 in animal tissues and excretions

Table 8 presents typical data on the levels of iodine 131 found in the milk, urine, feces and thyroid gland of the cow and goat following single dose and chronic administration.

The values are rough averages and the ranges are given in parenthesis. The variability is observed to be high and may be the result of biological differences between the individual animals and the sensitivity of iodine metabolism to the many variables operating. The most important values for food chain transference purposes are the milk concentrations. Typical values for the cow are 8 percent of a single dose in the total milk after seven days and approximately 1 percent of the daily dose per liter. The milk concentration variability, especially for the goat, is in part a result of the different levels of milk production. This is especially true for the goat. In estimating iodine 131 intake from milk, allowance must be made for the lesser absolute intake of iodine 131 by the goat. That is, produced on the same pasture, the concentration of iodine 131 in goat's milk would be 5 to 10 times that in cow's milk; however, the cow would most likely have a higher absolute quantity of iodine 131 in milk due to greater intake and milk production. Greater than 90 percent of the iodine 131 occurring in goat and cow's milk has been shown to be in the iodide form.

Another metabolic area of interest is the tissue distribution of iodine 131 as a function of time and type of intake. Typical data are presented in Table 9.

The important observation of Table 9 is the large difference between thyroid and other tissues. The difference is the result of the method of iodine 131 administration rather than the species. Tissue levels such as those of Table 9 could possibly be used to predict the tissue iodine 131 content of meat-producing animals under transient or long-term contamination conditions.

c) Factors modifying iodine 131 metabolism

Several investigators have reported a seasonal difference in the ability of the cow to secrete iodine 131 into milk.(16, 24) The magnitude of the difference and the mechanism operating are not known. It is probably an important physiological mechanism, and more work in this area is needed to determine the controlling mechanism(s). For public health purposes, this information is very important since it has an influence on how much iodine 131 is secreted into the milk. Along the same line of investigation, there is little evidence to illustrate a relationship between milk production and efficiency of secretion of ingested iodine 131 into milk. However, the total amount of iodine 131 secreted into milk appears to be a function of the amount produced with the quantity secreted per liter being relatively insensitive to changes in milk production.

Another area of radioiodine metabolism that has received considerable attention is that of adding various chemical compounds to the animal's diet in order to reduce the secretion of iodine 131 into milk. At the same time, the resulting reduction in thyroid burden of the animal has also been observed in hope of obtaining some indication of iodine behavior in the human population. Table 10 illustrates the effect of various chemicals on the amount of iodine 131 secreted into milk.

It is noted that the cows receiving no chemicals have an ability to concentrate iodines from blood plasma into milk. Little is known about the mechanism on how this is accomplished. With respect to this concentrating mechanism, the cow shows little ability when compared to the human being, dog, cat or guinea pig. Milk-plasma ratios in the cow range from about 1-4 while the other species mentioned have ratios of 20-30. The addition of chemicals is observed to reduce the milk-plasma ratio and the percent of the daily dose secreted per liter of milk for all cases (Table 10). However the reduction of the percent of iodine 131 secreted into the milk is not as much as would be predicted from the milk-plasma ratios. This is because the chemicals increase the plasma level of iodine 131 as well as reduce the milk concentration.

Table 8.(24) Secretions of Iodine 131 into the Milk, Urine, Feces and Thyroid of the Cow and Goat

Percent of single dose of ^{131}I*	**Cow**	**Goat**
In Milk**	8(4-20)	40(6-54)
In Urine**	50(30-75)	33(8-52)
In Feces**	20(13-50)	15(2-20)
In Thyroid	10(5-30)	30(2-50)
Percent of daily ingested ^{131}I	**Cow**	**Goat**
Per liter of milk	1(0.5-2.7)	65(22-150)
In Daily Urine	(35-50)	(10-50)
In Daily Feces	(20-40)	(10-30)
In Thyroid	(30-270)	(90-500)

*Dose refers to total intake activity

**All excretion values are cumulative totals over a 7 day period.

Table 9.(16) Tissue Distribution of Iodine 131 in the Cow and Sheep

	Cow	Sheep
Tissue	Single dose of ^{131}I[a] as percentage of dose/kg	Continuous dose of ^{131}I[b] as percentage of daily dose/kg
Thyroid	450	30,000
Muscle	0.02	-
Heart	-	2
Liver	0.03	8
Kidney	0.05	6
Lung	0.05	4
Pancreas	0.04	3
Blood	0.07	3

[a] Sacrificed after 7 days

[b] Sacrificed after steady-state conditions

Table 10.(16) Effect of Various Chemicals on the Concentration of Iodine 131 in Cow's Milk

Chemical	Amount g/day	^{131}I in milk as percentage of daily dose/l	^{131}I milk/plasma ratio
	0	2.3	1.40
NaI	10	0.7	0.27
	0	0.5	2.09
$NaSCN$	10	0.2	0.66
	0	1.5	2.6
$KClO_4$	10	0.4	0.32

Table 11.(24) Relation of Daily Intake of Stable Iodine to Stable Iodine Milk Concentration

Daily intake of iodine by cow (mg)	Iodine level in milk (mg/l)
1.6	0.028
20	0.27
2,000	19
4,000	25

Table 12.(24) Relative Biological Availability of ^{131}I to Individuals

Age	Thyroid wt., g	%uptake in thyroid	%uptake/g of thyroid	Fresh milk consumption	(%^{131}I/g thyroid) X(liters milk/day)
FETUS					
12 wk		0	0		0
12-15			0.1-1	1	0.1-1
15-32			1-5	1	1-5
PERSON					
0-6 mo.	2	30	15	0-0.5	0-7.5
6-12 mo.	2	30	15	0.5	7.5
1-2 yr.	2-5	30	12	0.5	6
2-5 yr.	3-5	30	10-6	0.5	5-3
5-10 yr.	5-10	30	6-3	0.7	4-2
10-15 yr.	10-15	30	3-2	0.7	2-1
15-20 yr.	15-20	30	2-1.5	0.6	1
20-30 yr.	20	30	1.5	0.3	0.4
Above 30	20	30	1.5	0.2	0.3

Another interesting sidelight of feeding chemicals to the cow is that the chemicals may be secreted into the iodine metabolism of the consumer. The data of Table 11 illustrate the daily intakes and milk concentrations of stable iodine for the dairy cow.

Assuming that man consumes about 0.2 mg of iodine a day, it is apparent that supplementing the cow's diet could significantly raise the iodine intake of individuals consuming milk. This metabolic behavior could have significant meaning (other factors being considered) in terms of public health action with respect to thyroidal deposition of iodine 131.

3. Metabolism in man

a) General behavior

Like radioactive strontium, radioiodine is a potential hazard to man since it is concentrated in significant quantities by the thyroid gland. The thyroid is a small, essential endocrine gland located in the neck and has a particular need for iodine. The gland synthesis iodine containing hormones which regulate the overall metabolism of the body. It ranges in weight from less than a gram at birth to about 25 grams in a 25 year old adult, remaining constant thereafter. As with all metabolic processes, very specific conditions must be defined in order to describe thyroid behavior since so many parameters take part in the process.

Iodine (inorganic iodide) is essentially 100 percent absorbed from the gastrointestinal tract. The thyroid gland traps from 10-50 percent of the absorbed amount, and oxidizes inorganic iodide to elemental iodine. The iodine may then be incorporated into the thyroid protein thyroglobulin. Enzymatic breakdown of thyroglobulin with subsequent reactions produce thyroxine and iodothyroxines, the active thyroid hormones. The hormones are secreted from the thyroid, circulated in the blood stream and are finally broken down. The free iodine may be retrapped or excreted. Excretion is mainly through the urinary route.

b) Biological availability

From a practical viewpoint, a relative factor expressing the biological significance of iodine 131 to the general population from food chain contamination can be expressed as (%^{131}I uptake per gram of thyroid) X (fresh milk consumed)(4). Table 12 presents several pertinent parameters from predicting the biological availability of iodine 131 in relation to thyroid burden.

It should be pointed out that the data of Table 12 are quite variable and single values are only used for illustrative purposes. Very little data are available for the uptake and thyroid weights of the fetus and young age groups. Limited studies on adults have shown that the uptake value may be somewhat less than that presently recommended (30 percent). The important point of Table 12 is that due to the combined effects of thyroid weight and milk intake the 0 to 2 year age group would be expected to have the highest iodine 131 exposure. The other groups would progressively lower exposures by factors of 10 - 20 at the ends of the age range.

c) Dietary supplementation

From the standpoint of possible practical application there is considerable interest in the effects of stable iodine and thyroid extracts on iodine 131 uptake. It is believed that the quantity of iodine retained by the thyroid from day to day is almost the same and is a function of the percentage of the total iodine intake which goes to the gland.(26) Sudden changes in stable iodine intake have little effect on changing the percent iodine uptake by the thyroid; however, chronic changes in intake causes the thyroid to adapt by large changes in the percent of absorbed iodine going to the thyroid. Studies have shown reduction of 75 percent or more of the iodine 131 accumulation in the thyroid with increased levels of stable dietary iodine.(26) Administration of thyroid extract has also been shown to have a similar effect.

C. Radiocesium

The major terrestrial pathways of cesium 137 are shown in Figure 4.

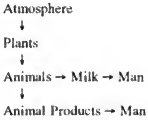

Figure 4. Major Terrestrial Pathways in the Food Chain for Radiocesium(5)

In contrast to strontium 90, cesium 137 does not readily follow the soil step. This nuclide is fixed in forms largely unavailable to plants as a result of entrapment in the lattice structure of certain clays. Plant contamination, therefore, occurs primarily by direct deposition. Since cesium 137 is capable of concentrating in soft tissues (e.g., muscle), the step from animal to animal products (other than dairy products) assumes importance for this radionuclide. A long-term genetic dose is thus possible from ingestion of foods contaminated with cesium 137. Since a significant quantity of cesium 137 is excreted into animal milk, the pathway atmosphere-plants-animals-milk-man is most important for diets containing average amounts of milk. In general, the biological significance of cesium 137 is somewhat less than that of strontium 90 because of its shorter effective half-life in the body.

Cesium 137 behavior in the food chain and concentration in various media is frequently expressed in terms of cesium-potassium ratios in a fashion analogous to that used for strontium and calcium. However, while strontium and calcium are closely interrelated in their food chain movement and can be used with relatively good degree of accuracy for prediction purposes, this is apparently not true for the cesium-potassium pair. Studies investigating the OR relationship at various levels in the food chain indicate that the various OR values change markedly under different conditions.(15) It appears that all of the weaknesses of the strontium-calcium ratio are magnified in the cesium-potassium pair. However, due to usage, the cesium 137 contents of diets and animals including man are often expressed as picocuries of cesium 137 per gram of potassium. This expression of concentration has some merit over that of other methods (e.g., picocuries per kilograms of body weight) since it correlates better with lean body mass, and hence organ dose, than does more conventional units. The concentration ratio may be approximately converted to other useful units since the potassium content of the body and many foodstuffs are relatively constant and well known. The ratio concentration also tends to minimize differences due to age and sex when these population characteristics are not known from the measured group. Use of picocuries of cesium 137 per gram of potassium in this fashion implies no consideration of the similarity of the metabolism of cesium 137 and potassium.

1. Behavior in soils and plants

The behavior of cesium 137 in soils and plants has not been studied to the extent of that of strontium 90. It is interesting to contrast the behavior of strontium 90 and cesium 137 since they are the major radionuclides of concern in evaluating long term environmental contamination. Where possible, this is the approach used in the following discussion.

a) Plant absorption and soil movement of cesium 137

The uptake of cesium 137 from soil has been studied in laboratory experiments and to a lesser degree in field experiments. These may be used for the basis of predicting field behavior. Cesium 137 has been shown to be less absorbed by plants than strontium 90. The relative amounts absorbed being dependent on the soil type. Short-term experiments indicate that cesium 137 from soils of the temperate regions usually enter plants to about one-tenth the extent of strontium 90.(15) With the passage of time, on the order of several years,the value may decrease to one-twenty-fifth of that of strontium 90. However, differences have been observed with other soil types. For example, tropical soils containing little clay minerals have shown greater plant uptake of cesium 137. The marked differences in the plant uptake of strontium 90 and cesium 137 from temperate soils is a result of fixation of cesium 137 by the soil in forms inaccessible to plants. The fixation appears to involve the entrapment of cesium 137 in the lattice structures of clay minerals. Once fixed, cesium is replaced from soil only to a small extent by divalent ions, however, addition of stable cesium, ammonium, or potassium reduces the amount of fixation. Addition of stable carrier cesium can increase plant uptake of cesium 137. Addition of stable potassium may decrease plant absorption of cesium 137 when the soil is low in available potassium. When available potassium is high no affect may be observed.

A typical study illustrating the comparative downward movement of cesium 137 and strontium 90 in several undisturbed soils is shown in Table 13.

It should be noted that the data of Table 13 are only valid in undisturbed soils since agricultural land which has been plowed and harrowed will have a top layer with a more homogeneous distribution of the radionuclides. As a result of strong binding (fixation) the downward movement of cesium 137 is considerably less than strontium 90 for the clay and sandy soils.

b) Direct contamination of plants by cesium 137

Cesium, like potassium, is readily absorbed and distributed throughout plant tissue. Studies performed to determine the metabolic interaction of cesium and potassium in plants are hampered by the appearance of toxic effects at relatively high ratios of cesium to potassium. However, studies investigating the OR (plant/solution) relationship have shown that the OR may be increased by a factor of 2-10 as the soil solution potassium concentration is increased.(15) This behavior points out the difficulty in trying to use the cesium-potassium ratio to describe the differential behavior of the two ions.

Cesium 137 may enter plants directly by foliar, floral or plant-base absorption. It is retained on plant foliage in a manner similar to that of strontium and is readily removed by rain. Like strontium 90, the relative rate of entry of cesium 137 into the plant tissues is not known. Once deposited on foliage, the cesium 137 is distributed throughout the plant. This is in contrast to radiostrontium which undergoes little distribution within the plant. For example, approximately 30 percent of the deposited cesium 137 on the foliage of potatoes may reach the tubers as compared with less than 1 percent of strontium 89.(17)

The extent to which cesium 137 enters plans from direct contamination as opposed to absorption from the soil has received considerable attention. Since large quantities of cesium 137 are fixed in various soils, plant levels would be expected to be closely related to the rate of deposition. In general, this idea is borne out (dietary surveys); however, in some regions the cesium 137 has not decreased in the expected manner with decreasing rates of deposition. Two possible explanations have been proposed for this observation.(17) First, cesium 137 undergoing plant-base absorption may be held available at the base of the plant for absorption for longer periods than currently anticipated. This may be particularly true for the root-mats of permanent pastures. Secondly, the slow rate of fixation may render additional quantities of the total soil reservoir of cesium 137 available for root absorption. More study in this area is needed before the relationships between the level of cesium 137 in plants, the cumulative soil deposit and the rate of deposition are fully understood.

2. Metabolism in domestic farm animals

Like radiostrontium, barium 140, and iodine 131, milk is the major pathway by which cesium 137 enters the human population. Although cesium 137 may be found in most tissues of the body, the soft tissue is generally considered the critical organ. The major area of study in cesium 137 animal metabolism has been the transfer of the radionuclide to milk. However, for cesium 137 this area has received less attention than for radiostrontium. Other metabolic characteristics which have received consideration are the qualitative and quantitative tissue distribution after single and chronic exposures, and the effect of dietary modifications on cesium 137 uptake.

Table 13.[16] **Comparative Downward Movement of Cesium 137* and Strontium 90 in Several Undisturbed Soils**

	Clay Soil		Sandy Soil		Calcareous soil**	
Depth(cm)	pH 4.6		pH 6.6		pH 7.6	
	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
0-2.5	50	98	63	98	37	43
2.5-5.0	20	2	22	2	24	21
5.0-7.5	15	0	13	0	19	15
7.5-10.0	12	0	2	0	16	16
10.0-12.5	3	0	0	0	3	4
12.5-15.0	0	0	0	0	1	1

*Percentage in each zone after 20 months experimental study
**Calcium carbonate containing

a) Secretion into milk of cesium 137

Studies have shown that about 10 percent of a single oral dose of cesium 137 will be secreted in the milk of the dairy cow in a 7 day collection period.[16] Daily administration of cesium 137 for long periods (30 days) results in about 10 percent of the daily dose being secreted into the milk. However, the animals of the particular study had not quite reached equilibrium with their intake. The amount of cesium 137 secreted per liter averages about 1.4 percent of the daily administration. These data from a typical study are presented in Table 14.

Table 14.[16] **Distribution of Radiocesium after Oral Administration to Dairy Cows and Goats**

Material	Cow	Goat
Milk (% daily dose)	13	7.0
Milk (% daily dose/l)	1.4	9.4
Urine (% daily dose)	30	40
Feces (% daily dose)	55	40

The relative order of distribution of the dose is the important concept of Table 14. It is noted that fecal excretion is more in the cow than in the goat. Also, a greater amount of cesium is recovered per liter of goat's milk. This is similar to the situation for radiostrontium and radioiodine. Radiocesium is excreted into the cow's milk by a factor of 10 greater than for radiostrontium. This is probably due to the greater gastrointestinal absorption of cesium than strontium. Body burdens of about 4 to 5 times the daily intake for the cow and goats, respectively, have been observed from chronic administration. Biological half-lives for the cow and goat have been estimated to be 20, and 2 to 3 days, respectively. Addition of stable cesium to the cow's diet has been reported as having little effect on the distribution of cesium 137 in the milk, urine, or feces.

b) Tissue distribution of cesium 137

The tissue distribution of a single dose of radiocesium to sheep and beef cattle has been observed to be similar.

Table 15 presents a typical 7 day tissue distribution of cesium 137 on several domestic animals after a single oral dose.

The concentrations of cesium 137 in the various tissues is seen to be of the same order of magnitude, kidney, showing the highest concentration. Since cesium 137 is a long half-life radionuclide, its tissue distribution after chronic ingestion is more applicable to long term environmental contamination. The data of Table 16 show the tissue distribution of cesium 137 in the goat as a result of chronic feeding.

The various tissues show a reasonably similar quantity of radioactivity. The kidney appears somewhat high while the brain is rather low. Comparison with Table 15 indicates that other species (cow, sheep, etc.) would be expected to have tissue distribution on the order of magnitude of that of the goat when exposed to chronic intake.

Table 16 also presents several calculated observed ratios for the cesium-potassium pair in the tissues of the goat. In this particular study a OR value of 1.3 indicates that the goat preferentially secretes radiocesium into the milk. A similar value (1.6) has been reported for the dairy cow. For the organs and tissues of Table 16 with an observed ratio greater than one (liver, lung, kidney, and mammary gland), a concentration of cesium 137 with respect to the diet is implied. This suggests that cesium does not follow potassium metabolisms in the manner and closeness that strontium follows calcium metabolism. Further investigation of cesium-potassium metabolism is needed before any definite conclusions can be stated. For example, limited studies in rats indicate that a ninefold increase in dietary potassium causes a twofold reduction in cesium 137 muscle retention. (Table 17).

The OR(muscle/diet) is observed to vary as a function of potassium content of the diet; a ninefold increase in diet carrying a fivefold increase in the OR(muscle/diet). This is in direct contrast to the predicted behavior of the OR(bone/diet) for strontium.

Table 15.(16) Radiocesium in the Tissues of Farm Animals after an Oral Dose* (percentages)

Tissue	Cow	Sheep	Pig
Blood	2.8	4.7	6.0
Plasma	1.3	2.6	1.0
Muscle	30	41	23
Liver	26	24	21
Kidney	45	74	26
Femur Chaft	0.5	7.6	5.2

*Data are expressed as percentage of dose x 10^5/g, corrected to 500 pounds body weight

Table 16.[16] Equilibrium Distribution of Radiocesium in the Tissues of the Goat*

Tissue	Cs^{137} as percentage of daily dose/kg	Cs/K Observed Ratio (tissue/diet)
Muscle	4.6	0.6
Brain	0.5	0.3
Liver	8.4	1.4
Lung	7.3	1.1
Kidney	17.0	3.7
Mammary Gland	7.1	1.4
Blood	0.5	0.6
Milk	5.6	1.3

*25 days of feeding

3. Metabolism in man

Cesium 137 is absorbed from the gastrointestinal tract of man virtually completely and primarily distributed to the soft tissues of the body. There has been some evidence that significant quantities of cesium 137 are concentrated in bone; however, results at present are quite variable and further investigation is needed before any definite conclusions can be drawn. Cesium 137 like potassium has a relatively short biological half-life in man and equilibrium with the intake is established rather rapidly, regardless of age.

At least two exponential processes (2 compartment model) can be recognized in the elimination of cesium 137 from man following the administration of a single dose.(15) A small fraction (10 to 15 percent) is excreted with a biological half-life of 1 to 1.5 days. The remainder is eliminated at a much slower rate with considerable variation between individuals and biological half-lives. Values as low as 50 to 60 days and as high as 150 to 160 days have been reported for the biological half-life. Measurements on apparently normal individuals with whole body counting techniques have yielded an average value of 100 days. Potassium, on the other hand, has a much more rapid body turnover which apparently depends largely on the dietary intake.

Table 17.(19) Effect of Dietary Potassium on Cesium 137 Retention in Muscle of Rat*

Level of Dietary K%	**%Daily Dose of muscle Cs^{137}**		**Dietary Cs/K %Daily Dose/mg**	**$\frac{Cs/K}{Cs/K^{muscle}}$**
	per gram	per mg K		(diet)
0.2	14	3.9	3.9	1.0
0.6	10	2.9	1.3	2.2
1.8	7.5	2.1	0.4	5.3

The biological half-life of cesium 137 in different species appears to be related to body size - being rapid in small animals like the mouse and rat, and longer in large individuals such as man. Ruminants (e.g., cattle) form an exception to the rule. They have a rather rapid biological half-life when compared to similar sized animals. The level of dietary potassium appears to have little effect on the removal of cesium 137 from the human body. An increase of dietary potassium causes only a transitory increase in the urinary excretion of cesium 137, and it shortly returns to its former rate.

D. Other Radionuclides

Several additional radionuclides which are either man-made or natural constituents of the terrestrial environmental deserve brief consideration.

1. Fission and activation products

Radionuclides produced in the fission process, other than those of the alkali and alkaline earth groups, will generally be of minor importance because they are short-lived and pass through terrestrial food chains to only a small extent. None of these radionuclides are absorbed by man in any appreciable amount.

In addition to the fission products, a wide variety of induced activities may result from the activation of material surrounding the fission reaction or through the release of effluent from reactors. Radionuclides originating from these situations include isotopes of iron, sodium, manganese, magnesium, phosphorus, cobalt and zinc. None of these radioisotopes enter the terrestrial food chain to any extent approaching that of the contaminants already described. However, several of these radionuclides (e.g., phosphorus 32 and zinc 65) concentrate in aquatic forms and may reach man through the aquatic food chain.

Carbon 14 (half-life 5560 years), both natural and man-made, is produced from atmospheric nitrogen. Its natural production is the result of cosmic ray interaction; man-made carbon 14 is produced from interaction of neutrons, with nitrogen. Past atmospheric weapons testing (up until 1959) has increased the natural occurring carbon 14 level in the troposphere of the Norther Hemisphere by about 30 percent.[17] Carbon 14 readily enters the biological carbon cycle and contributes to the whole body population dose. Tritium (hydrogen 3, half-life 12.5 years) like carbon 14 is both naturally occurring and man-made. Natural production is again by cosmic ray interaction; man-made production occurs by activation of atmospheric nitrogen by neutrons coming primarily from weapons testing. Tritium can also be reactor-produced by neutron activation of deuterium (heavy water) moderator and lithium hydroxide, a coolant additive. Natural tritium levels have been modified by atmospheric nuclear weapons tests, especially by high yield fusion devices. As expected both carbon 14 and hydrogen 3 are readily incorporated into all media of the food chain.

2. Natural radioactivity

There are a number of naturally occurring radionuclides in the biosphere. They enter and are transferred through the food chain to varying extents. A major portion of the work with natural occurring radionuclides has been undertaken in order to establish base-line levels in the environment. Such levels have been used for comparison in assessing the degree of contamination of the environment with artificial radioactivity.

The most important naturally occurring radionuclides belong to the uranium, thorium and actinium series. The actinium series is the least abundant of the three. These natural series are composed of a number of the heavier elements of varying half-lives which exhibit extremely complex decay schemes. Studies of the transfer of these radionuclides to man via the food chain involves measuring the activity levels in plants which are consumed directly by man, and plants such as grasses which form the principal food of animals which in turn become the principal food of man. Dietary surveys indicate that there is a wide range of natural activities in vegetation and there appears to be no simple correlation with the activities found in the soil which have a much smaller range. Also, there is not much information available concerning the discrimination factors for soil-food and man-food processes. It appears that the majority of these radionuclides are absorbed by plants and animals only to a very small extent when compared to the most pertinent artificial radioisotopes. Little quantitative data are available on absorption phenomena. The greatest attention has been paid to radium since this element appears to be the principal one absorbed by plants. However, dietary surveys have indicated that the occurrence of this element in the majority of foods is well below that of man-made strontium 90.

The following naturally occurring radionuclides are present in a singular form in the environment: rubidium, lanthanium, samarium and ruthenium. Little information is available on plant and animal absorption of these natural radionuclides. They are known to be present in the environment at considerable lower concentrations than the most abundant naturally occurring radioisotope, potassium 40. Consideration of several of their characteristics such as their chemical nature, the behavior of their man-made counterparts and their essentialness to plants would tend to indicate that these radionuclides are little absorbed by plants.

SUMMARY

In order to determine the environmental significance to man of a particular artificial radionuclide many characteristics of the environment and man must be considered. Such characteristics include the fission-yield and half-life of the radionuclide, the rate of entry and survival of the contaminant through the terrestrial food chain, the rate and quantity of uptake of the radioisotope by terrestrial media (plants, animals, man, etc.), the amount of the contaminant deposited and retained in the critical organ of man, and the type and energy of the radiation emitted by the radioisotope. Additional physical and bio-chemical characteristics of the radionuclide must be considered and related to the contaminant's behavior in individual terrestrial food chain media. From the above considerations, strontium 90 and cesium 137 are the most hazardous long-term contaminants that can readily reach man through the terrestrial food chain. Strontium 89, barium 140 and iodine 131 are also potential internal hazards to man; however, as a result of their shorter physical half-lives they are most significant as relatively short-term contaminants.

Much work has shown that the most important terrestrial pathway in the food chain for these radionuclides is atmosphere-plant-animal-milk-man. In most cases this is the pathway by which these radionuclides reach man the quickest and in greatest quantity. Other pathways in the food chain may assume significant importance as contributors, depending on the particular situation.

The remaining fission products, activation products and naturally occurring radionuclides that are found in the terrestrial environment are of lesser hazard to man as a result of their limited movement in the food chain. However, depending on special circumstances some of these radionuclides may assume a greater importance than normally expected.

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b) Radionuclides in the Food Chain

INTRODUCTION

The use of nuclear energy by man involves the production of artificial radionuclides which present a potential public health problem with regard to the contamination of the terrestrial food chain. The release of these radionuclides has not introduced to our environment a new pollutant but rather has increased an old pollutant in terms or radiation exposure. The problem then is a matter of evaluating the extent of man-made radioactivity in the environment and its subsequent health significance.

At present fallout from past nuclear weapons tests is the most widespread source of artificial radionuclide contamination to the terrestrial environment. With limited nuclear testing other sources such as, nuclear reactor operations, nuclear fuel reprocessing, waste disposal and medical and industrial uses of radioisotopes will be receiving more emphasis as significant sources of environmental contamination.

This discussion is primarily concerned with the qualitative behavior and transfer of atmospheric releases of man-made radionuclides through the food chain and their deposition in the human population. Where applicable reference is made to the behavior of the more important naturally occurring radionuclides of the uranium, thorium and actinium series along with the singularly occurring radionuclides.

THE FOOD CHAIN

The pathways which radionuclides follow in moving from their origin to man constitute the food chain.

Radioactive materials are removed from the atmosphere by meteorological processes, primarily precipitation. In general, the most serious food chain contamination problem arises from direct deposition of radioactive materials on animal feed crops or on food crops directly consumed by man. Following this initial deposition, the radioactive materials may be removed by various processes such as, being washed off by rain or blown off by wind. The extent of this removal is a function of many physical and biological parameters. Man's intake of radioactive material may occur from contaminated food crops, from contaminated meat and meat products and from contaminated milk or milk products. The inhalation route (atmosphere directly to man) may be important under special circumstances. The relative importance of the various pathways of intake will depend on many factors among which are the physical half-lives of the radionuclides, the rate and route by which they pass through the food chain and the dietary habits of the population. The immediate and generally most significant pathway is pasture-cow-milk-man for the more significant radionuclides up to approximately one hundred days following deposition (one time event). Plant losses are such that after this time period adsorption from the soil by plants is the most significant pathway for the longer-lived radionuclides. The final step in the food chain (uptake by man) primarily depends on the chemical characteristics of the radionuclide and the metabolism of the concentrating organ.(2)

As an example of radionuclide behavior in the terrestrial food chain, the pathway of the fission product strontium-90 may be cited. It has been shown that this nuclide is readily transferred to man from atmospheric nuclear testing by deposition on the earth's surface with subsequent plant uptake and transference to milk by the grazing animal population.

In considering the relationships between the food chain and any particular radionuclide it becomes necessary to first consider the source of the contaminant. The source should be considered at it relates to the physical and chemical state of the radionuclide since these properties relate to the degree of movement in the food chain. Fission-products from an atmospheric burst would not be expected to exist in the same physical and chemical state as those from a reactor incident, fuel reprocessing plant or waste treatment facility. Even among the latter operations a wide variety of physical and chemical states of the fission-products would be expected.

As an example of the foregoing the various forms of the fission products of ruthenium and iodine with respect to their source of production can be cited.

Studies of the movement and effects of radionuclides in the food chain are of value because they 1) make possible the prediction of the relationships between the kinds of radioactivity in the various steps within the food chain and the resulting levels in the human population, 2) provide a means for evaluating ways by which the levels in the human population may be minimized, 3) provide background information for setting up environmental sampling programs and for interpreting the data obtained, 4) make available animal data which may be useful for estimating behavior of radionuclides in humans, and 5) make possible predictions of the possible effects of environmental contamination, especially in the grazing animal population.

RADIONUCLIDE BEHAVIOR IN TERRESTRIAL MEDIA

Radioactive materials from atmospheric releases are deposited on the earth's surface by precipitation or direct deposition. Although the discussion of possible fallout patterns is beyond the scope of this presentation, it should be stated that fallout distribution depends on many parameters. For nuclear weapons testing, these include meteorological conditions, fission yield, type of explosion (i.e., ground, water or air), and geographical location. In the case of a nuclear incident, such as an air release from a nuclear reactor, the factors of major concern would be the existing fission product inventory, the micrometeorological conditions and the nature of the surrounding terrain.

The incorporation of radionuclides from fallout into the food chain depends on the physical properties of the fallout material and the chemical behavior of the nuclides. The observed wide range of chemical composition and particle sizes in various nuclear events leads to the conclusion that the biological availability of the radioisotopes cannot be predicted from the event without some knowledge of the characteristics cited above. With respect to weapons testing, surface bursts in the kiloton range over continental soils result in predominantly siliceous radioactive particles while tower bursts of the same energy range yield particles which exhibit incorporation of tower materials. Megaton explosions over coral islands have yielded primarily calcareous (calcite or calcium carbonate containing) particles.(3)

Very little quantitative information is available on the exact physical and chemical form of the metallic radionuclides that constitute the bulk of the radioactivity from world-wide fallout. The information that is available has been obtained from the various methods of sampling fallout as it impinges on the earth's surface. Comparisons have been made using gummed films, pots, ion exchange columns connected to funnels, and collectors employing artificial grass. It has been observed that gummed surfaces collect on the average about 60-70 percent more gross radioactivity per unit of horizontal area than a high walled pot. The alkali metals (e.g., cesium-137) and alkaline earths (e.g., strontium-89 and strontium-90) are collected at a much lower efficiency on gummed surfaces than the less reactive metals and their less soluble oxides (e.g., zirconium-95, cerium-144 and yttrium-91). The lower collection efficiency for the presumed soluble fallout (alkali and alkaline earth metals) has been attributed to washing off by rain. When rain water is collected in a pot and filtered, nearly all the strontium, barium and cesium isotopes pass through the filter and are presumed to be in solution at the time they are deposited in rain (this may not always be true for cesium). Conversely, the less reactive material composing the bulk of the gross radioactivity is generally associated with the particulate material.(4)

A. Soil-Plant Considerations

For discussion purposes, the movement of radionuclides through the food chain can be conveniently considered by examining the system's major components. These are the soil, plant and animal. However, in order to evaluate the overall hazard from deposition and movement in the food chain all the components must be considered on an interrelated behavioral basis.

A major part of the biological experimentation concerned with radionuclide movement in the food chain, especially with fallout constituents, has been done with soluble forms of the respective radioisotopes. Consequently, the observed effects are assumed to exceed those which would be expected from the less soluble fallout. Use of soluble sources is presumed to provide maximal effects.

1. Soil considerations(5)

Radionuclides presently in soils are of two kinds - natural and artificial. The natural radionuclides are either extremely long-lived such as potassium-40, thorium-232 and uranium-238, or they are daughters of long-lived radionuclides such as radium-226, polonium-210, bismuth-212, etc. The artificial radionuclides are man-made fission products which have become widely distributed in soils by atmospheric circulation and deposition. The most important radionuclides of this group being those of relatively long half-life such as, strontium-90 and cesium-137.

Although the natural radioactivity contents of soils is much higher than that of the fission products, the movement of natural radionuclides in the food chain has not caused much concern for human health. Possible reasons are that these radionuclides have always been with us and natural variations in their abundance have not caused any apparent differences in human well being. Except for the element potassium and radium, there have been few studies on the availability of natural radionuclides in soils for food chain transference. For potassium the majority of studies carried out have been more concerned with the natural elements' nutritional value rather than its radioactive significance. On the other hand, there has been much concern for fission products in soils of agricultural importance and in materials from these soils that are used for food. This concern is due primarily to 1) the artificial nature of these fission products, 2) their potential as significant contaminants of the food chain, 3) the health implications to the human population, and 4) a need to be able to predict the behavior of much higher concentrations of them in soils. Much work is presently being undertaken to find ways to minimize plant uptake of these fission products from soils. This area of study can be termed the effects of "soil amendments".

a) Soil characteristics(6)

Soils consist of mineral and organic matter, water, and air arranged in a physicochemical system that provides for plant rooting and nutritional supplement. A vertical profile through various soils reveals horizontal layers which differ in their physical characteristics and their ability to support different kinds and quantities of vegetation. In a typical situation three layers may be recognized. The uppermost, which may range from 1 to almost 2 feet in thickness, is the surface soil which supports the majority of the life processes taking place. The second layer consists of the subsoil, extending to about three feet below the surface. Further below the surface yet, to a depth of about 5 feet, is a layer of loose and partly decomposed rock which is the parent material of the soil.

The inorganic portion of the surface soil may be categorized into a number of textural classes, depending on the percentage of sand, silt and clay. Sand consists predominantly of primary minerals such as quartz and has a particle size ranging from 50 microns to about 2 millimeters; silt consists of particles in the range of 2 to 50 microns; while, the clay particles are smaller than 2 microns in diameter. The clay fraction of soils is largely responsible for controlling the processes which provide nourishment for plants. The platelike particles of aluminum silicates comprising the clay contain an abundance of negative surface charges which attract positively charged ions. This is one of the most important properties of soils. Most of the positively charged nutrient ions in soils are not in the water phase, but are absorbed on the surfaces of soil particles. In this way a greater reservoir of nutrients can be held in solution by the soil water. If soil clays did not have this ability to bind cations many dissolved nutrients would not remain in soils but would be leached from them by normal water movement. In contrast to cation nutrients in soils, anion nutrients generally exist in combination with organic matter or as anions while in the inorganic form. These anion nutrients must generally undergo decomposition to inorganic anion forms before being utilized by plants. There is no anion exchange mechanism in the soil similar to the cation exchange mechanism.

Cations entering soils in water solution are exchanged with cations adsorbed on the surface of clays. With time soils become acidic as a result of the replacement of adsorbed cations by the hydrogen ions of water. Because of this hydrogen ion buildup soils must be limed (addition of calcium hydroxide) from time to time to assure optimum plant growth. The liming procedure replaces the hydrogen ions with calcium and magnesium ions.

b) Soil reactions(5, 6, 7, 8, 9, 10, 11)

Soil contamination is the first step in the process of plant root uptake. Radionuclides are made available for uptake by 1) soil penetration by rain water or natural erosion processes, and 2) mixing into the soil by cultivation practices. In the first case, the resulting concentration gradient of radionuclides coupled with the rooting pattern and depth of plant structures are important considerations in plant uptake. In the second case, cultivation methods such as plowing, harrowing and discing mixes the fallout to depth of cultivation and increases the uptake by plants grown in soil. In general rapid soil penetration is dependent on the structure of the soil, adsorptive properties of the radionuclide, electrolyte concentration of the soil and the rate of water movement down through the soil. Rapid penetration is enhanced by low humus and low clay content, low valence, high electrolyte concentration of the soil and rapid movement of water.

Since the adsorption of nutrients or inorganic ions by the roots of plants usually involves soluble exchangeable ions in the soil, the radioactive ions compete with and replace other ions on exchange sites in the soil. In some of the reactions with the soil, the new ions of fallout origin become nonexchangeable and some quantity of the radionuclides become unavailable for plant uptake. Thus, the availability of a radionuclide for plant uptake from the soil depends on the types of interactions that occur between the soluble radionuclide and the soil constituents. Several of these soil radionuclide interactions are listed below.

Adsorption(6). The exchange capacity is the term commonly used to designate the ability of a soil to adsorb cations. It is usually expressed as the number of milliequivalents of cation(s) required to neutralize the negative charge of 100 grams of soil at a given pH (usually 7). In general, the adsorption of an ion will depend on its ion exchange properties-valence, ionic size and chemical form in solution. Radionuclides exhibiting favorable ion exchange properties will be removed from the soil solution in a sequential manner dependent on their relative affinities for the adsorption media. For example, a trivalent ion would be more readily adsorbed than a divalent ion which in turn would be more readily adsorbed than a monovalent ion. However, other factors may override the above. Thus, cesium-137 (monovalent) is more strongly adsorbed than strontium-90 (divalent) in many soils as a result of its entrapment in the lattice structure of some clays.

Desorption(3). Desorption can be considered in its broadest sense as the reverse of the adsorption process. Those radionuclides which are most strongly adsorbed will be the most difficult to desorb or leach from the soil. In the strontium-cesium example cited above, water desorption of cesium is less than that of strontium. The amount of desorption of the above two ions can be altered by the characteristics of the leaching solution (salt concentration, acidity and presence of complexing agents) and by the characteristics of the soil (base saturation and buffer capacity). Addition of amendments such as lime and organic matter also change desorption phenomena. Since adsorption is the primary and initial retention mechanisms in the soil and desorption is the primary mechanism affecting availability to the plant root, both adsorption and desorption phenomena are important in considering subsequent plant uptake. These parameters are especially important with respect to the rate and total quantity of radionuclide movement in the soil.

Presence of other ions(3). The presence of other ions exert a strong effect on the adsorption of a particular cation. When used in large amounts all cations will reduce the adsorption of strontium and cesium. The usual replacement of cations on soil materials is lithium < sodium < potassium < ammonium < rubidium < cesium < hydrogen < magnesium < calcium < strontium < barium < iron < aluminum < lanthanium. The presence of anions have also been shown to have a marked effect on cation adsorption. The order of decreasing strontium adsorption in the presence of anions is sulfate > chloride > nitrate while increased adsorption occurs with oxalates and phosphates.

Effect of pH(3). Most studies of pH effects on soil reactions have varied the pH of the leaching solution. These studies have attempted to duplicate the actual condition which is variation of soil pH instead of pH variation of the contaminating solution. Such investigations have yielded data of the type illustrated in Table 1.

Table 1.(3) **Maximum Adsorption as a Function of pH**

Radionuclide	**pH**
Strontium	7-9
Cesium	>6
Yttrium	>6
Cerium	>6
Plutonium	2.5-9

At highly acidic and alkaline conditions, soil minerals decompose and yield decomposition products which compete with the fission products for adsorptive sites. Under these conditions it is expected that there would be less radionuclide adsorption.

Fixation(3,9). Fixation of radionuclides in the soil is a process that renders these constituents unavailable to plants. Neutral salt solutions are usually used to extract the exchangeable cations from soil. These extracted cations are considered to be the cations that are available for plant nutrition. The amount of fixation in a particular soil is the difference between the amount of cation applied and the available or extractable amount. The degree of fixation is an arbitrary term which depends on the experimental methods used. Current investigation in this area has indicated, especially with respect to strontium and cesium, that fixation is a property of the soil type since both fixed and non-fixed quantities of these elements have been found in soils. Several mechanisms that have been proposed for the fixation process include precipitation as slightly soluble materials, physical trapping between clay platelets and in other insoluble precipitates, and diffusion into existing crystals.

Effect of organic matter(10,11). Additions of large quantities of organic matter (tons per acre) affects soil reactions and subsequent plant uptake. The major factors involved here appear to be an increased microbial activity which competes with the soil and plant for the mineral elements; and an increased tie-up of ions from the soil solution by the adsorptive capacity of the added organic matter. The net effect observed with respect to radionuclide behavior is a reduction in plant uptake. The amount of reduction is primarily a function of soil type and radionuclide.

2. Plant characteristics

Plants are a major vector in the transfer of radionuclides from the environment to man. Contamination occurs by two processes: soil uptake and aerial uptake.

a) Soil uptake(3, 6, 12, 13)

Although almost every element can be found in soil to some degree, sixteen elements are considered necessary for the growth and reproduction of plants. These are carbon, hydrogen, oxygen, nitrogen, phosphorus, sulfur, potassium, calcium, magnesium, iron, manganese, zinc, copper, molybdenum, boron and chlorine. Carbon, hydrogen and oxygen are primarily supplied by the atmosphere while all others are supplied from the soil. Radionuclides of the above elements will pass into the plant root in the same manner as the stable forms. In some cases the radionuclide element may not be required for normal metabolism (e.g., iodine, cobalt, uranium and radium); however, they are known to be present in plants although no metabolic function is known. In general it can be assumed that if the ion is in the soil it will probably occur to some greater or lesser extent in the tissue of the plant grown in the soil.

The more important factors that affect the uptake of radionuclides by plants through their root systems are 1) the chemical and physical form of the ion of the radionuclide, 2) the plant species and its metabolic requirements, 3) the soil type and chemical characteristics and 4) the soil management practices. Numerous theories have been proposed for the mechanism of uptake of ions by plant roots. Most hypothesis agree that the uptake of the element occurs in an ionic form, and the concentrating of the element within the plant root is a metabolic function. Most of the mechanisms also provide for a biological compound that serves as a carrier for the element. Disagreement in the various hypothesis occurs in the areas of the exact mechanism of uptake and the transporting mechanism(s) of ions between soil and plant root.

The relative uptake of various elements from soils have been shown to be strontium > iodine > barium > cesium, ruthenium > cerium > yttrium, promethium, zirconium, niobium > plutonium.(12) Where commas appear between the isotopes, difficulty has been encountered in determining the relative order of magnitude of uptake. In a particular soil at comparable activity levels the relative order or uptake by plants does not vary much but the total quantity taken varies considerably with different soils. Variations that are encountered in the order and absolute magnitude of uptake result from the differences in radionuclide concentrations in the soil and in the reactions of the various radionuclides with the different soils. With respect to radionuclides from nuclear detonations, about two percent of the radioactivity in a contaminated soil may be absorbed by a single crop of plants.(3) However, soils usually adsorb less than a tenth of a percent of the total deposited. Strontium-89 and strontium-90 absorption may account for as much as seventy percent of this total from one year old mixed fission products. It is generally accepted that about one percent of the applied strontium will be taken up by single crops of plants. Less than one tenth of a percent of the deposited amount of other elements is taken up by single crops of plants. Higher amounts of uptake of some of the radionuclides may be obtained in pot experiments.

The translocation of radionuclides is believed to be primarily governed by the uptake process and less by plant metabolism. The main route appears to be the central zone of the vascular tissue of the plant. Upward movement of the elements strontium and calcium relative to that of phosphorous, sulfur, iodine and rubidium is limited. Cesium, rubidium and potassium exhibit a greater redistribution within the plant than does strontium, calcium, yttrium and other multivalent cations.(3,12) Radionuclides of higher valence such as strontium-90, ruthenium-106, cerium-141, yttrium-91 and zirconium + niobium-95 tend to accumulate in the leaves to the highest extent relative to fruits and stems. Radionuclides of lower valence such as cesium-137, rubidium-86 and potassium-40 are distributed more uniformly throughout the plant and exhibit less preference for a particular plant structure. Plant content of these radionuclides will vary with the stage of plant development. With respect to species differences, the order of fission product uptake by the different plant families has been reported as Leguminasae (pea family - clover, alfalfa, beans, etc.) > Solanaceae (potato family - tomatoes, potatoes, tobacco, etc.) > Compositae (composite family - dandelions, lettuce, sagebrush, etc.) > Gramineae (grass family - crab grass, wheat, rice, etc.) for the tops and Leguminasae > Compositae > Solanaceae for the roots. However, other investigators report no consistent differences between the lower and higher plant families.(3)

b) Aerial contamination and uptake(3,6,8,13,14,15,16)

In addition to indirect plant contamination (i.e., absorption from the soil) three methods of direct plant contamination have been established:

Foliar deposition. This term refers to the deposition of radioactive materials on the leaves of the plant. The deposited radioactive material may a) adhere to the leaf surface, b) be absorbed into the leaf or c) washed from the plant onto the soil. The magnitude of plant contamination from this process will depend on the radionuclide(s) deposited, plant species and existing weather conditions.

Floral absorption. This term refers to the absorption of the deposited materials by the inflorescences (flowering parts) of plants. For example, the inflorescences of wheat plants have a shape that tends to maximize entrapment of fallout particles. Thus, contamination of this plant shortly before maturity results in greater radionuclide absorption in the peripheral part of the plant.(8)

Plant base absorption. This term describes the absorption of material that has been washed down to the basal parts of the plant. Material absorbed in this fashion may or may not be diluted by other elements of the soil.

Plant contamination may thus be caused directly by foliar and floral contamination or indirectly by absorption from the soil. A combination of direct and indirect contamination is possible by plant-base absorption.

The relative importance of these alternative routes of entry will depend on the growth form of the plants and the role of the various parts of the plant in relation to the dietary habits of man. Foliar absorption occurs readily in all species whereas floral absorption is of practical significance with grain crops only. Plant-base absorption is particularly important in perennial pastures but not necessarily confined to them. In many cases it is difficult to distinguish between foliar absorption and either floral or plant-base entry. During rain the contamination may wash downward over the surfaces of the plant and foliar and floral absorption merge into plant-base entry. As materials are further leached into the soil surrounding the plant-base, plant-base entry becomes indirect absorption through the roots.

Direct contamination is also a function of the size and solubility of the contaminant. Large particles characteristic of near-in fallout from nuclear detonations usually rebound from leaf surfaces. In general particles larger than 40 microns in diameter are only slightly retained.(16) Particles larger than this diameter are associated with local fallout but are not characteristic of fallout at considerable distances from the detonation. Insoluble materials which are more characteristic of near-in fallout result only in superficial plant contamination. The greatest plant contamination occurs from finely divided soluble deposits - world-wide fallout is in this form. It is important to note that findings based on one situation are not applicable to the other (i.e., plant contamination due to local versus world-wide fallout).

Little data are available showing the extent of fission product uptake by above-ground plant parts from world-wide fallout. However, evidence is increasing that the major portion of plant contamination from fallout has resulted from deposition on foliage.(8,16) The exact quantity contributed may primarily depend on the plant's growing habits and on the existing weather conditions during the absorption period. Tracer studies indicate that entry can occur through the leaves, stem, fruit and bark. The extent of entrance of materials in solution into plants appears to be directly proportional to the amount applied and to the time period of absorption. Other significant information such as the rate of absorption, extent of internal transport and pattern of distribution within plants is dependent on the specific radionuclide, plant and other internal and external factors. With respect to the extent of internal transport, several studies have indicated both acropetal (transport to the tip or extremities of the petal) and basipetal (transport to base of petal), transport occurs following application of phosphorous-32, potassium-42, rubidium-86 and cesium-137 to above-ground plant parts. In contrast to this calcium-45, strontium-89 and 90, yttrium-90, ruthenium-103 and barium-140 do not move freely from the absorbing organ and basipetal transport is negligible.(15)

3. Soil-plant relations(3,13)

The soil and plant components of the soil-plant system are individually complex. The system is dynamic in that there is a steady input of ions by the soil and a steady uptake of ions by the soil and a steady uptake of ions by the plant. When the input equals the uptake for a specific element steady state conditions prevail. From a nutritional standpoint, steady-state conditions are probably common for many vegetative forms in the environment. However, for world-wide fallout this is most probably not the case because of the limited residence time of most radionuclides in the soil. The degree of attainment of steady-state conditions may often be regulated by one or more of the soil-plant factors mentioned in the preceding sections.

a) Effects of soil amendments

Various soil amendments have been investigated in order to reduce plant uptake of radionuclides from the soil. Among these are lime, gypsum, fertilizers and various types of organic matter. Most work in this area has been concerned with the fission-product strontium-90 since it is the most hazardous radionuclide that undergoes absorption from the soil. Limited work has been reported for cesium-137, ruthenium-103 and cerium-144.(3)

When amendments are added to the soil two processes occurring within the soil are altered: 1) the exchange reactions governing the distribution of ions between the clay and the soil solution and 2) the competitive effects during the course of absorption by plants. Since various cations will compete for the same adsorption site, increasing the concentration of one decreases the uptake of others in the same group. This hypothesis has been proved to be correct within certain ranges by numerous greenhouse and field experiments. For example, the addition of calcium to acid soils low in calcium content results in a reduction of strontium-90 uptake. Similar observations have been observed on the uptake of cesium-137 from soils low in potassium when stable potassium is added. In some investigations fertilizer has increased the uptake of fission-products, but in others no increase occurred. The effects of organic matter have been cited previously.

In general, the soil that will provide minimum uptake of fission-products appears to be one that is ideal for maximum crop growth. These prerequisites include a soil of high exchangeable calcium, high exchangeable potassium, high organic matter content and a slightly alkaline reaction.

b) Concentration factors and observed ratios(3)

Since the uptake of a radioelement depends on its concentration in the external medium, the term concentration factor is often used to express the relative uptake of different elements. It is defined as the ratio of plant tissue concentration to the external medium concentration. Results from solution culture studies are usually expressed in the fresh tissue weight while oven dry weight is used for soil culture studies. The formula for calculating concentration factors is presented in Equation 1.

$$\text{C.F.} = \frac{\mu\text{Ci/g plant tissue}}{\mu\text{Ci/g external medium}} \tag{1}$$

There is relative agreement in the order of concentration factors for different radioisotopes using various laboratory and field procedures. Some reported concentration factors for fallout radionuclides in soil culture experiments are 0.05 for the alkaline earth group and 0.009 for the rare earths. Using soluble forms in nutrient solution concentration factors from 0.05 to 1.0 have been observed for strontium, cesium, iodine and barium. For ruthenium, yttrium and cerium the range is 0.0001 to 0.001. Thus, the concentration factor is an arbitrary term which depends on the experimental conditions.

Another term used quite often to describe the quantitative soil-plant relations is the Observed Ratio. The OR is used to report the uptake of fission-products relative to a chemically similar element. For example, the OR for strontium is the ratio of strontium to calcium in the plant(sample) divided by the ratio of strontium to calcium in the nutrient medium (precursor). For calculation purposes the Observed Ratio is determined as indicated in Equation 2.

$$\text{OR(sample/precursor)} = \frac{\text{Sr/Ca(sample)}}{\text{Sr/Ca(precursor)}} \tag{2}$$

The OR (plant/nutrient solution) for strontium relative to calcium when only plant discrimination is measured is close to 1.0. This indicates no discrimination against strontium and is generally true for all plant parts except the roots, where OR values larger than 1 have been observed. Other OR values reported for the rubidium/potassium and cesium/potassium systems in various plants are 0.85 and 0.20 respectively. These systems indicate discrimination against rubidium and cesium in favor of potassium.

c) Soil versus aerial contamination(2,17)

In evaluating the effect of deposition to man an additional consideration of some importance in soil relations is the significance of soil versus aerial contamination with respect to subsequent transfer of radioactive materials up the food chain. More recent deposition is believed to contribute predominantly to foliar and floral contamination (direct), whereas the older deposit regulates the absorption from the soil. If contamination of man's diet comes primarily from the direct mechanism, the radioactivity levels in the diet would be expected to be related to the quantity of current deposition. Under these conditions a decrease in the rate of deposition would result in a rather rapid decrease in dietary contamination. Alternatively, the primary mode of dietary contamination may be the indirect method. In this case the levels appearing in the diet would be dependent on the cumulative deposit in the soil and the extent of equilibrium conditions within the soil system. Since the cumulative deposit and attainment of equilibrium conditions are time dependent, dietary contributions from soil absorption would be of greater significance on a longer term basis. If deposition ceased completely, dietary levels resulting from soil absorption would fall slowly due to the decreasing availability of fission products to plant forms. This decreased availability would be due to 1) the slow radioactive decay of the cumulative deposit and 2) the loss of existing soil deposited radionuclides through physical processes (e.g., leaching).

B. Plant-Animal Considerations

From the preceding sections, root uptake and aerial contamination have been identified as the methods by which radioactive materials contaminate crops that are eaten by man or that serve as food for stock animals. Much remains to be learned about how each radionuclide moves from the root into the edible portion of the plant, through the body of animals and into the milk, meat and other foods of man. More yet remains to be learned about the behavior and distribution of radionuclides in the bodies of animals and man. A quantitative understanding of these relationships is needed before the dose received by man from the deposition of a radionuclide in the environment or the concentration in a food can be evaluated.

1. Contamination of animals and animal products(8)

Certain radionuclides are readily transferred to the human population via domestic grazing animals which are effective collectors of contamination from various vegetative forms. There are many factors which effect the degree to which animals are contaminated. The most important include 1) pasture type used for grazing, 2) extent of barn feeding (purchased and stored), 3) miscellaneous feeding practices - age of food and supplemental feeding use of purchased feed, etc., 4) water source and 5) animal housing - degree of sheltering animals from surface contaminants. Because of the many variables involved the degree of animal and animal product contamination may be quite variable, even from apparently similar sources.

Of particular interest in this pathway of the food chain are the relationships that exist between the quantities of radionuclides ingested by the animal and the subsequent quantities which are deposited in the tissues and secretions that serve as human food. To study these relationships requires a knowledge of the metabolic characteristics of the animal and the particular radionuclide. The following section on radionuclide metabolism presents a general discussion of various aspects of the subject.

2. Radionuclide metabolism(18,19)

Classically, metabolism refers to the biological processes whereby complex cellular elements are synthesized (catabolism) or conversely complex substances are degraded (anabolism) to supply energy for biochemical reactions. Kinetics usually refers to the phases of metabolism involving rates of transfer and compartment volumes, i.e., various tissue spaces into which a substance is distributed. In general, from the point of view of radiological hazards, interest focuses mainly on the kinetic aspects of metabolism.

Radionuclide metabolism is arbitrarily considered as organic or inorganic though such a distinction cannot, in fact, be made. Organic metabolism is somewhat synonymous with the more commonly referred to processes of intermediary metabolism. Illustrative of this would be the passage of carbon-14 through a biological system. Minerals, e.g., potassium, sodium, calcium, strontium, etc., may be viewed as being an inorganic phase. This arbitrary separation only simplifies the study of radionuclide metabolism.

a) Experimental methodology in metabolic studies

Metabolic studies of radionuclides and oftentimes their stable counterparts are applications of tracer experimentation. Tracer concepts can generally be used for the study of radionuclides in biological systems though one must realize the assumptions inherent in the theory. Illustrative of the latter are situations in which a tracer is not homogeneously distributed in a compartment such as the skeleton.

Only fundamental approaches will be cited. Upon introduction of isotopes into the animal body, concentrations in various body fluids, excretory products and biopsies containing assimilated material can be analyzed. Sacrifice of the animal and proper dissection lead to more precise distribution and concentration assays. Techniques of autoradiography lead to knowledge of differential distribution as well as quantitative data. Yet, methods employing sacrifice give data only applicable to some increment in time.

A method of nondestructive study would be ideal but such is not yet available. In recent years, techniques of total body counting have been developed and, applicable in localizing as well as quantifying radioisotope within the body. At present, this system is only useful in studying gamma emitting nuclides within the animal.

Most metabolic data have resulted from single dose experiments. In some instances, single administration data may not provide complete information and multiple doses of the same nuclide may be required. Study of skeletal metabolism is probably the best example of the latter since a non-homogeneous distribution results from a single dose. More exacting procedures utilizing multiple radioactive forms of the same nuclide can greatly enhance the information in a study of metabolism. Thus chronic administration, especially simulating fallout conditions, needs to be carried out to give an adequate picture of the metabolism of certain nuclides.

b) Methods of data interpretation

In the study of radionuclide metabolism most data have been represented by an exponential function with regard to accumulation, transfer and elimination:

$$R = Ae^{-\lambda t} \tag{3}$$

where:

R = quantity in biological compartment at time t
A,λ = constants characteristic of the compartment
e = exponential function

The latter is appealing and would be expected if the biological system functioned as a simple diffusion process. Also, the concept is similar to the exponential decrease seen with radioactive disintegration. Hence the concept of biological half-life analogous to radiological half-live evolves and one may speak of a biological half-life for loss of the radionuclide through biological processes. With more detailed metabolic models, a multiple exponential form has been used. This model assumes that the radionuclide is distributed among several biological compartments and is lost from each with a characteristic half-life. The equation takes the form:

$$R = Ae^{\lambda_1 t} + Be^{\lambda_2 t} + Ce^{\lambda_3 t} + \ldots \tag{4}$$

where:

R is the amount retained at time t and A, B, C and λ_1, λ_2 and λ_3 are constants characteristic of the various compartments.

Another expression that has been used quite often relates retention of some radionuclides, especially bone seekers, to a power function of time:

$$R = A_t^{-b} \tag{5}$$

where:

R is the amount retained at time t and A and b are constants.

With a sufficient number of terms, the multiple exponential expression can give as good a fit to experimental data as the power function. However, when both methods are extrapolated beyond the data on which they are based the two forms lead to widely varying results. It is probable that a number of mathematical functions will ultimately be required to adequately interpret the multiple biological functions inherent in metabolic phenomena.

c) General considerations involved in radionuclide metabolism

1. Routes of assimilation

For practical purposes only the gastrointestinal mode of entry of nuclides into the compartments of a biological system are important. In special situations, the pulmonary and skin routes may be important in permitting assimilation. The intravenous route of entry is artificial and only important as an experimental tool. However, it must be noted that it simulates the situation once a nuclide is absorbed into the blood stream. This method to some extent bypasses the uncertainties involved in study with natural routes of assimilation.

The gastrointestinal system is probably the most important route of entry for soluble forms of nuclides. Insoluble forms will, dependent on the degree of solubility, remain in the gut with this organ receiving the bulk of the radiation exposure. Alpha emitters will dissipate about one percent of their energy in the tissues of the gastrointestinal tract. A much greater percentage of the energy from beta emitters will be absorbed and dissipated within the gastrointestinal tract. Insoluble gamma emitters will effectively radiate the intestine, however, the rest of the body will also be exposed.

Pertinent examples of the more important fission products are tabulated in Table 2.

Table 2.(8,20,21) Characteristics of the More Important Fission-Products of Food Chain Significance

	Yield				
Radioisotope	U^{235}(a) (%)	U^{238}(b) (%)	Type of radiation	Physical half-life	GI absorption(c) (%)
Sr^{90}/y^{90}	5.1	3.2	β	28 years	30
Cs^{137}	6.2	6.2	β, γ	29 years	100
Pm^{147}	2.9		β	2.6 years	0.01
Ce^{144}	5.0	4.5	β, γ	285 days	0.01
Ru^{106}/Rh^{106}	0.5	2.7	β, γ	1.0 years	0.03
Zr^{95}	6.3	5.7	β, γ	65 days	0.01
Sr^{89}	4.6	2.0	β	51 days	30
Ru^{103}	3.4		β, γ	39.7 days	0.03
Nb^{95}	6.3		β, γ	35 days	0.01
Ce^{141}	6.0		β, γ	33 days	0.01
Ba^{140}/La^{140}	6.1	5.7	β, γ	12.8 days	5
I^{131}	3.1		β, γ	8.05 days	100

(a) Thermal neutron fissioning, (b) Fast neutron fissioning, (c) Assume radionuclide occurs in soluble form

Gastrointestinal absorption for the nuclides range from 100 percent for iodine-131 and cesium-137 to less than one percent for seven of the other potential environmental contaminants listed. Table 2 assumes that the radionuclides occur in a soluble form on ingestion. Most are both gamma and beta emitters. Active versus passive absorptive mechanisms plus the physicochemical nature of the nuclide are responsible for selective absorption. Yet, nuclides such as iodine-131 are 100 percent absorbed, no matter what form in which they are administered. Absorption of strontium can vary markedly, dependent upon chemical forms whereas almost all forms of zirconium are insoluble.

2. Solubility versus insolubility

The physicochemical nature of the radionuclide bears great influence on its passage through a biological system. Such factors as the particular compound in which the nuclide is incorporated, the valance state and ionic form, particle size, complex formation, and whether bound to a protein make great differences in the biological activity of radionuclides. These parameters are usually simplified by categorizing the physicochemical form of the nuclide as soluble or insoluble. Also, it is important to recognize that some nuclides especially those high in atomic number are insoluble of themselves, regardless of any other physical or chemical characteristic. Thus, often a barrier to the incorporation of certain nuclides into biological systems exists due to their very nature.

3. Influence of other nuclides on assimilation

The presence of other elements and compounds of other elements will often greatly influence the assimilation of nuclides. Numerous interrelationships exist between metabolic phenomena, e.g., influence of calcium on strontium retention.

4. Absolute versus relative uptake

In certain instances, especially due to the nature of the biological mechanism involved, there may exist relative and absolute degrees of nuclide assimilation. Certain biological systems may have specific quantitative requirements per unit time. For instance, would calcium continue to be absorbed from the diet if its concentration in the diet was constantly increased? More likely an absolute amount may be absorbed per unit time with rejection of the excess dietary calcium. If assimilation were to be expressed in terms of percent absorption in this instance, one would be misled. Thus, the concept of relating assimilation to fractional uptake could be inaccurate in describing the biological phenomena.

5. Specific activities and biological availability

The expression of nuclides in terms of specific activity (SA = radioactive/stable) often times more adequately describes the biological processes. However, it must be stipulated that the radioactive and stable forms of the nuclide be homogeneously mixed before specific activities of various compartments may be intercompared. In the diet, presence of the radioactive and stable elements as different compounds with different solubilities may make an expression of specific activity invalid, for the physicochemical form must be the same in order that both be metabolized in the same way. Hence, a specific activity may be present in the biological system, even after equilibrium, because of differences in physicochemical availability.

6. Deposition of nuclides within the biological system

Little generalization can be made about nuclides in this all important phase of metabolism. Nuclides do fall into somewhat gross categories such as "bone seekers", "marrow seekers", etc. Although a nuclide may have an inclination for certain organs it is important to realize that metabolism in that organ may be considerably different for different nuclides seeking the same organ. Thus, calcium and radium can be incorporated into bone in entirely different ways.

7. Routes of elimination

Urinary and fecal excretion are by far the most important exits of nuclides from the body, varying in importance with particular nuclides. Sweat, sloughing of various skin structures, exhalation and secretions of minor organs may contribute to excretion. Perspiration losses of calcium, sodium, and potassium may be significant and should be recognized in balance studies.

d) Metabolic classification of radionuclides

No completely satisfactory classification of nuclides is possible. One approach is to group the nuclides according to position in the periodic table. It would be expected that nuclides of the same group would behave similarly because of their like chemical properties. However, caution must be taken in accepting the latter assumption. As examples, elements of Group VII, fluorine, chlorine, bromine, iodine, and astatine have strikingly different modes of metabolism. Fluorine is deposited in bone, chlorine and bromine are fairly equally extensive with the extracellular fluid space, and iodine is concentrated in the thyroid gland. Astatine is also localized in the thyroid gland.

1. Hydrogen (tritium)

Tritium is of major importance as tritiated water. Tritium equilibrates rapidly with total body water and its loss from the body is determined by the turnover rate of the body water which can vary widely depending upon fluid intake. An average biological half-life of twelve days is assumed. Tritium oxide is one of the few radionuclides absorbed through the intact skin.

2. The noble gases

Of neon, argon, krypton, xenon and radon, only the latter has received considerable attention. The metabolic characteristics of this group is characterized by differential solubility in body fats and the absence of chemical reactions with body components. Radium is transferred through the food chain and deposited in bone with subsequent radon liberation. Recent developments in this area are concerned with the retention of the liberated radon. Recent investigation indicates that in long-standing cases of radium poisoning an average of seventy percent of the radon produced is exhaled.

3. The alkali metals - Group I

Though, sodium, potassium, rubidium and cesium are members of Group I, their metabolic fates are considerably different. Again, pointing out the fact that like chemical properties, does not always indicate like metabolism. Indeed, their biological half-lives are 11, 35, 80, and 110 days respectively - a marked range.

Potassium-40, the most abundant natural isotope in biological systems is not a fission product. Its exceedingly long physical half-life of years has resulted in an abundance of the radionuclide remaining since the time of creation. Its distribution is more principally intracellular in mammals with much lower concentrations present in extracellular fluids (lean body mass). It is one of the most easily measured gamma emitting radionuclides present in animals. Roughly, there are 2.2 grams of potassium per kilogram of lead body mass and it is one-hundred percent absorbed with most of it being excreted in urine.

Sodium is of interest mainly as an indicator of dose after neutron exposures with production of sodium-24 in the body. Studies with sodium-22 have elucidated the metabolism of this element. It is distributed mainly in the extracellular fluids with much lower intracellular concentrations. Significant concentrations of sodium are retained in the mineral portion of bone with a very slow turnover rate.

Cesium-137 and cesium-134 are fission products found as human contaminants. Cesium seems to be principally concentrated in muscle, but has a similar whole body distribution to that of potassium. The metabolic fate of rubidium more closely resemble cesium than the other alkali metals.

4. The transition elements of Group I

Copper, silver and gold have no immediate environmental importance as contaminants of the food chain. The common radioisotopes of these elements have relatively short half-lives, limited gastrointestinal absorption and are used primarily in medical applications.

5. The alkaline earths - Group II

As environmental contaminants, elements of this group are undoubtedly among the most important. Barium, lanthanium, radium, calcium, strontium and yttrium are probably the most intensively studied group of nuclides since they are readily transferred through the food chain to man. Strontium, barium and radium are metabolized in a manner similar to calcium. In the animal body they are rapidly and almost exclusively deposited in the skeleton. Some of the radionuclides of longer half-life deposited in the skeleton will remain there throughout life. A single exposure or a number of short term exposures result in a heterogeneous pattern of bone deposition. Chronic exposure results in a more uniform pattern of deposition. This dependence of distribution on duration of exposure further complicates dose evaluation.

Barium-140, a fairly short-lived fission product, is of lesser metabolic interest due to its short half-life (12.8 days) and limited gastrointestinal absorption. The metabolic behavior appears very similar to that of strontium.

Research into the metabolism of radium dates at least to the 1920's, stemming from industrial and medical contamination of many people with the radionuclide. Gastrointestinal absorption is approximately 30 percent of ingested dose. In general, radium is deposited in bone and is retained there for long periods of time. Irregular deposition in osteons with "hot spot" formation has been stressed resulting in tumor formation, etc., if the concentration is great enough. Normal total body concentration has been estimated at 1×10^{14} gram. The degree of deposition in other tissues of the body is unsettled and there is some confusion on this point with recent reports of high concentrations found in soft tissue. Recent results of long term studies suggest a much different retention for radium as compared to other bone seekers when interpreted by a power function. This point is extremely important, in that, regulatory standards of deposition of other radionuclides in bone have been based on radium metabolic data.

Radionuclides of the lighter alkaline earths, beryllium, magnesium and calcium have been used extensively in studies of their metabolic behavior but there is no particular environmental hazard from any of these isotopes.

6. The transition elements of Group II

Only zinc-65 is of interest as an environmental contaminant being transmitted in the food chain through aquatic biota exposed to reactor wastes. Neutron activation during fission results in its presence in close-in fallout. This radionuclide is said to be concentrated in the prostate and to a lesser degree in bone and skin, though it still has a wide spread distribution throughout the body.

7. Group III

The radioisotopes of boron, aluminum and scandium have attracted little interest as internal emitters. Gallium, indium, and thallium have been studied with respect to medical applications. Yttrium and lanthanium are very similar to the rare earths and are considered later with these elements.

8. Group IV

Carbon-14, a natural environmental contaminant due to cosmic ray activation of nitrogen-14, has been increased by about 10 percent through neutron activation of the atmospheric nitrogen-14 consequent to nuclear detonation. The metabolism of carbon is very complex and is almost synonymous with intermediary metabolism. Levels of carbon-14 in man and in the atmosphere (assuming no further nuclear testing) should drop off faster than anticipated from the carbon-14 half-life because of the continuing equilibration of new carbon-14 with the total biospheric carbon. Other elements of this group have been studied for their biological and therapeutic effects but little interest is attached to then as internal hazards from food chain transference.

9. Group V

Only phosphorous-32 is of major importance as an environmental contaminant. It is an activation product formed in reactor cooling water and can be concentrated to significant levels by aquatic food chain forms. Niobium as a daughter product of zirconium-95 is present in fallout materials but is of no more importance than its parent because of limited gastrointestinal absorption. Other radioisotopes of this group are of primary interest from a toxicological and biochemical aspect.

10. Group VI

There are no prominent fission product radionuclides in this group with respect to significant food chain contamination. Molybdenium-99, a fission product and chromium-51, an activation product, are introduced into the environment in limited quantities but are of little significance as a result of their relatively short half-lives and limited gastrointestinal absorption. Sulfur and polonium have been the most extensively studied isotopes of this group. Radioisotopes of the transition elements of this group, chromium, molybdenium and tungsten have all been used in biological studies.

11. Group VII

The isotopes of radioiodine are the only fission product in this group of significant environmental importance. They are characterized by their ability to concentrate in the thyroid gland and their short physical half-lives.

12. Group VIII

Although the activation products iron-59 and cobalt-60 may be released to the environment from reactor operations and associated processes, they are not environmentally significant contaminants because of their limited absorption in terrestrial organisms (mainly limited gastrointestinal absorption). However, they are used extensively as biological tracers in medical and related areas. Of the other elements of this group, only ruthenium has received considerable attention. Ruthenium-103 and 106 are prominent short-lived fission products resulting from nuclear testing. Though virtually non-absorbed by various food chain forms, the small amount gaining entry into animals is deposited initially in the kidney and liver with longer retention in the skeleton.

13. The rare earths and actinide elements

The rare earths (lanthanides) and the actinides in addition to yttrium behave metabolically similarly. The rare earths are produced in high yield from fission reactions of the elements of the actinide series. These radionuclides behave similarly in the majority of their chemical and biochemical reactions. Once absorbed most are deposited in bone with degrees of localization in the liver and reticuloendothelial system (that which engulfs foreign particles in the animal body). There is a noted progression with increasing atomic number in the chemical behavior within these groups (more noted in the lanthanides) which is reflected in changes in their biological behavior. With increasing atomic number, there is a greater tendency for bone deposition and less for deposition in the liver. The lighter elements tend more to fecal and heavier ones more toward urinary excretion. Bone retention is much longer than in other organs. Their distribution appears to be particularly sensitive to the quantity of material involved. Carrier free radioisotopes are concentrated to a greater or lesser extent in bone while, with the addition of stable element along with the radio-element, there is a tendency for deposition in the liver and reticuloendothelial system. The exact mechanism of bone deposition for these groups is believed to be different than that for the alkaline earths.

Although radionuclides of these elements are widely distributed over the earth, they have not been identified in appreciable quantity in plants, animals and man. This is a consequence of their extremely low solubility in biological materials and corresponding low absorption from the gastrointestinal tract. In most experimental animals, less than one-hundredth of a percent of an ingested dose is absorbed.

COMMENTARY

The contamination of the terrestrial food chain from the various uses of nuclear energy constitutes a potential health problem to man. To cope with this problem requires a consideration of the physical, chemical and biological characteristics of the radiocontaminants and the terrestrial food chain. The methods of fission product deposition along with the mechanisms of soil, plant and animal contamination need to be fully understood. With the above considerations in mind, the particular radionuclides of health importance to man in the terrestrial environmental can be identified and their metabolic relationships studied. By sampling the appropriate media a comparison of the quantities of these artificially produced fission products with the naturally occurring radionuclides can be made and then an assessment of the degree of environmental contamination can be obtained.

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II. METHODOLOGY

A. PREPARATION OF SAMPLES

Preparation of food samples for analysis of radionuclides involves three major operations. Each is essential to obtain results which are valid, accurate, and precise.

The first is to insure that the food sample represents the edible portion or the portion actually consumed. Inedible material such as bone, apple cores, nut shells, and egg shells should not ordinarily be included as part of the sample. To be valid as a dietary sample of a meal, the food for the entire meal should be prepared and "served" in reasonable portions or multiples thereof. Any convenient amount of a single food item can be sampled and the radionuclide content reported on a weight or volume basis.

The second operation in food sample preparation is to make the sample homogeneous so that aliquots will be identical in composition. This is best accomplished by blending or homogenizing using commercial blenders or specially constructed homogenizers, depending on the sample load and typical sample size.

The third operation is to put the homogeneous sample into solution. It is accomplished by ashing in a muffle furnace, digesting in acid, by fusing, or by a combination of these. The volatilization temperatures of the radioactive elements to be analyzed influences the choice of procedure. Volatile gamma emitters such as radioiodine and cesium isotopes can be measured in the undissolved homogenate by gamma spectrometry. If a radioelement, volatile at dry ashing temperatures, cannot be determined by gamma spectrometry, it can be analyzed chemically after the sample has been dissolved by acid digestion.

Fresh samples should be analyzed immediately upon receipt or refrigerated for a short time only or frozen. Food samples should be packaged in waterproof containers, and if frozen during storage, thawed prior to analysis. This treatment obviates the need for adding a preservative to the samples. Convenient sample and aliquot size for individual food items or composites of known constituents can be determined from the Table A.

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1. Blending

Several methods of sample blending may be used to homogenize liquid, semi-liquid, and solid samples of food, excluding inedible, hard and resistant parts such as bones and skin. One of the procedures described here can be applied to large or small number of samples or to samples of large or small volume. Because they differ primarily in the type of apparatus employed, choice of procedure will be dictated by the sample load, sample size, availability of funds and equipment, and personal preference of the analyst.

Principle of Method

The food sample is weighed and thoroughly blended in a homogenizing apparatus until the sample becomes a homogeneous slurry. Portions of the slurry are measured into appropriate containers for analysis by gamma spectrometry and/or for ashing prior to radiochemical analysis.

PROCEDURE

Reagents

none

Apparatus

evaporating dish
laboratory balance
Marinelli-type beaker, 3.5-liter capacity

stainless steel electric food blender, 1-gallon capacity

Procedure

1. Weigh the total sample, and homogenize it in a 1-gallon capacity blender. After the sample appears to be a slurry, blend it for 3 minutes longer.

2. Measure 3.5-liters of the homogenized sample into a tared Marinelli type beaker and determine the aliquot weight. This portion is counted for gamma activity in a gamma scintillation spectrometer. After the blended sample is counted for gamma activity it may be transferred to a pre-weighed evaporating dish to be ashed in preparation for further analysis.

2. Ashing

Two similar procedures for ashing food samples are given here. They can be used in connection with the method of blending described above or be applied to a blended dietary sample or a raw, cooked, or unhomogenized food item, although special care must be taken in ashing intact fatty substances. If not homogenized, most samples should be cut into small pieces before being ashed. In each procedure, ambient temperature is progressively increased to reduce the consistency of the sample to that of a coarse gravel.

Muffle furnaces are the only major laboratory equipment needed, and the number of samples that can be ashed per day depends primarily on the number and capacity of furnaces available. The minimum time required to ash a single sample is a little over three days; however, the sample needs to be watched or handled only occasionally.

Principle of Method

The blended sample is dried and then ashed in a muffle furnace by progressively increasing the furnace temperature to 500°-600°C. The ash is then cooled, weighed, pulverized or mixed by a similar method, and divided into portions needed for analysis.

PROCEDURE A

Reagents

none

Apparatus

Corning Ware dish
electric drying oven
evaporating dish
gas oven
muffle furnace
plastic container

Procedure

1. Transfer a suitable portion (see Table A) of a blended sample to a tared Corning Ware dish and obtain the aliquot weight. Dry the sample in an electric drying oven at 115° C for 48 hours.

2. Transfer the sample to a gas oven and char for 4 hours at 250° C. Then raise the temperature to 315° C for an additional 4 hours. Then raise the temperature to 480° C and char for 8 hours more. Remove the sample, and cool it.

3. Transfer the cooled, charred ash to an evaporating dish and place it in a vented muffle furnace. Raise the temperature to 550°-600°C and ash for 16 hours.

4. Cool, weigh, and then grind the ash to a fine powder using a mortar and pestle.

5. Transfer the powdered ash to a plastic container from which portions can be removed for chemical analysis.

PROCEDURE B

Reagents

none

Apparatus

Corning Ware dish
drying oven
muffle furnace
plastic container

Procedure

1. Transfer a suitable portion (see Table A) of a blended sample to a tared Corning Ware dish and obtain the aliquot weight.
2. Dry the sample in an oven at 100° to 120° C for 48 hours.
3. Transfer the sample to a vented muffle furnace, and raise the temperature slowly from 200° C to 550°C. Keep the temperature of the furnace at 550°C and ash the sample for 48 hours.
4. Cool, weigh, and then grind the ash to a fine powder using a mortar and pestle.
5. Transfer the powdered ash to a plastic container from which portions can be removed for chemical analysis.

3. Fusion

If a food sample contains a high concentration of silica, it may be necessary to perform a sodium carbonate fusion on the ash in order to dissolve it.

SODIUM CARBONATE FUSION

Principle of Method

Barium and strontium carriers are added to an aliquot of food ash and the mixture is fused with sodium hydroxide and sodium carbonate. The melt is taken up in distilled water, washed, and treated with sodium carbonate to yield a precipitate of alkaline earth carbonates. The carbonate precipitate can then be analyzed for strontium 89, strontium 90 and barium 140.

Reagents

Carrier solutions:
Ba^{+2} as barium nitrate, $Ba(NO_3)_2$: 20 mg Ba^{+2} per ml
Sr^{+2} as strontium nitrate, $Sr(NO^3)_2$: 20 mg Sr^{+2} per ml
sodium carbonate, Na_2CO_3: anhydrous granules or powder, 3N, 0.1N
sodium hydroxide, NaOH: pellets

Apparatus

long-handled tongs, nickel-tipped
nickel crucible, 250-ml capacity
nickel spatula

Procedure

1. Weigh a 10-gram or smaller portion of food ash into a 250-ml nickel crucible. Record the weight. [Note: Sample size depends on the silica content of the ash.]

2. Pipet 1.0 ml of strontium carrier solution and 1.0 ml of barium carrier solution (or an amount of carrier suitable to the analysis to be made) into the crucible. Mix thoroughly with a nickel spatula.

3. To this mixture add 50 grams of sodium hydroxide pellets, or 5 grams for each gram of ash, and mix thoroughly.

4. Fuse over a blast burner for 20-30 minutes. Then slowly stir in 5 grams of anhydrous sodium carbonate, or 0.5 gram for each gram of ash. Continue to heat the clear red melt for 20-30 minutes.

5. Using long-handled, nickel-tipped tongs, remove the crucible from the flame and very cautiously cool it in a cold-water bath. Take up the cooled melt in about 200 ml of hot distilled water and disintegrate it by gentle boiling.

6. Using distilled water, transfer the mixture to a 250-ml centrifuge bottle. Centrifuge, and discard the supernatant liquid.

7. To the residue add 40 ml 3N Na_2CO_3 as a wash. Heat for 10 minutes on a hot plate. Centrifuge, and discard the supernate.

8. Repeat Step 7 to put the precipitate in a form suitable for further analysis.

Table A. Required amount and approximate ash content of various sample media

Medium	Required amount (kg)*	Approximate ash content (%)
Apples	13.0	0.3
Bananas	4.0	0.9
Beans (green, snap)	5.5	0.6
(kidney, dry)	2.5	1.6
(large lima, dry)	2.5	1.6
(pinto, dry)	2.5	1.8
Beef (meat)	5.0	18.0
Bread	1.7	2.0
Cabbage	7.0	0.5
Celery	4.5	0.8
Cereals (All Bran)	0.5	8.2
(Cheerios)	1.0	4.5
(Corn Flakes)	1.5	3.1
(Grape-Nuts)	1.5	2.6
(Rice Krispies)	1.5	3.1
(Shredded Wheat)	2.5	1.4
(Special "K")	1.5	3.0
(Wheaties)	1.0	4.8
Coffee (instant)	0.4	12.0
Crab (whole)	0.5	14.5
DietFood (composite)	3.5	1.0
Eggplant	5.0	0.8
Eggs	5.0	0.7
Fish (whole)	0.5	6.5
(edible, red snapper)	2.0	1.8
Grass (dry)	1.0	5.5
(green)	2.0	2.0
Lettuce	7.0	0.5
Milk (evaporated)	2.5	1.6
(fresh)	5.0	0.7
Margarine	1.6	2.3
Oranges	9.0	0.4
Oysters (meat)	0.5	6.0
Pork (meat)	5.0	0.8
Potatoes (fresh)	4.0	1.0
Pumpkins	8.5	0.4
Rice (white)	7.0	0.5
Rutabagas	6.0	0.6
Shrimp (whole)	1.0	4.5
Squash	5.0	0.7
Tomatoes	4.5	0.8
Watermelon	13.0	0.3

*In order to have sufficient ash for the chemical procedures listed in this manual.

B. INSTRUMENTATION

1. Laboratory Facilities

The physical location of the room and the arrangement of the equipment are important factors to be considered when establishing a counting facility.

The site should be chosen so as to take maximum advantage of any natural existing shielding. By locating the room in the basement of a building it is generally possible to utilize natural shielding as well as to obtain maximum separation from busy areas. Temperature, humidity, and dust control are all important and equipment should be provided to optimize and stabilize these environmental parameters.

Cleanliness and general good housekeeping procedures are of the utmost importance in the operation of a low level counting room. Such practices tend to assure minimum background levels by reducing or eliminating contamination and this leads to more accurate results. Traffic in and out of the counting room should be kept at an absolute minimum. This may be accomplished by the use of in-wall sliding glass door pass boxes for the submission of samples for processing. It is often an excellent idea to provide shoe coverings for personnel entering the room.

2. Instruments for Measuring Radioactivity

Instruments for measuring radioactivity make use of the ionizing properties of nuclear radiation. Basically they provide a medium with which the radiation can interact and a means of detecting and identifying this interaction. All instruments can be classified by their interaction medium under three general categories: gas ionization detectors, scintillation detectors or phosphors, and solid state detectors.

When a charged particle such as an alpha or beta particle moves through a gas at high velocity it may act on a gas molecule or atom to remove an electron and produce an ion pair. Gas ionization detectors contain an enclosed volume of gas in which two collecting electrodes are placed. When a direct voltage is applied across these electrodes in the presence of ionizing radiation, five types of response will be observed as the voltage is increased. These five distinct regions of response are:

a In the ion recombination region many of the ion pairs recombine with each other before reaching the collecting electrode. Gas ionization instruments are not operated in this region of response.

b In the ionization chamber region the applied voltage is sufficient to prevent significant recombination of ion pairs and the ions are collected in a one-to-one ratio with those formed. Ionization chamber instruments operate in this region of response.

c At voltages in the proportional region the ions are accelerated to achieve enough energy to produce secondary ionization in the gas medium, thus amplifying the primary ion current. Although the ion current or pulse size is amplified, to an extent described by the "gas amplification factor", it is still proportional to the energy of the incident particle or radiation. Thus gas ionization detectors operating in the proportional region can detect alpha particles selectively in the presence of beta (or gamma) radiation. Alpha and beta proportional counters with or without windows to separate samples from the gas ionization medium operate at voltages in the proportional region of response.

d Geiger counters, or GM counters, operate in the Geiger-Mueller region where the gas amplification factor is so large that an avalanche of electrons spreads along the entire length of the anode. Here the pulse size is independent of the number of primary ions or energy of the incident radiation. This accounts for the high sensitivity of GM detectors and their inability to distinguish between the various types of radiation.

e In the continuous discharge region the gas amplification factor is so large that the gas begins to arc generating a series of self-perpetuating discharges. Operation of any gas ionization instrument at voltages in this region even for a few seconds will seriously damage the electrodes.

Scintillation detectors make use of the long-known ability of ionizing radiations to produce short-lived flashes of light (scintillations) in phosphors. The light flashes can produce photoelectrons from a photosensitive cathode in a photomultiplier tube and these photoelectrons are further amplified into a pulse which can be counted. When the scintillator is large enough to absorb all the energy of the radiation in the excitation and ionization interaction, the output pulse will be proportional to the energy of the incident radiation. This proportional response is necessary for gamma scintillation spectrometry. Scintillators are of many materials including organic crystals such as anthracene, liquid solutions of an organic scintillator such as p-terphenyl in an organic solvent, solid solutions of an organic scintillator in a plastic solvent (plastic scintillation detectors), inorganic crystals such as sodium iodide or zinc sulfide, and noble gases.

Solid state detectors make use of the property of some insulators such as diamond or silver chloride crystals, or semiconductors such as germanium or silicon, to show an instantaneous conductivity when high-energy particles or radiation interact with the material of which they are composed.

C. DETECTORS

a. Alpha Activity

Alpha particles are helium nuclei and carry a positive charge equal to twice the electron (beta) charge. Alpha particles, like gamma rays, are monoenergetic emissions. The most commonly counted alpha emitters, such as radon and thoron daughters, have energies of 5 to 7 million electron volts (MeV) and a mean range in air of about 5 centimeters. Samples to be counted for these particles should be very thin to minimize loss of energy in the sample through self-absorption, and should little or no distance between the sample and the detector to minimize loss of energy in air. Because of the low penetrating power of alpha particles, end-window counters are generally unsatisfactory for alpha counting unless the window thickness is 0.1 milligram per square centimeter or less.

For each instrument described, there is an operating high voltage which will yield consistent counting results. To determine the operating voltage, counts are made with a known alpha source at 50-volt increments. The initial counts will appear at the threshold voltage of the instrument, the lowest at which pulses are detected. As the voltage is increased, more counts are observed, until a leveling-off of the count rate occurs. This level area, or plateau, continues until there is a sharp increase in the counts. The operating voltage is selected midway along the plateau. The manual provided with each commercially available instrument usually discusses in some detail the selection of the probable operating voltage.

After the operating voltage has been selected, calibration must be performed to obtain quantitative data. Calibration involves selecting a suitable standard, preparing it in a form to simulate the samples, and counting the calibration standard at the selected operating voltage.

For calibrating alpha counters, sample thickness must be considered because of self-absorption in the sample can be severe. Performance standards are a very necessary part of routine counting. Their use insures that the counter is functioning the same when the sample is counted as it was when calibration was performed.

Background count rate should be determined at least twice daily to detect possible fluctuations caused by contamination, radio-frequency or line-current pulses, or by a change in the natural background radiation from radon-thoron daughters in the air or from cosmic radiation. A change in background due to contamination of the counter is most apt to occur if the samples are placed within the detector for counting.

1. INTERNAL PROPORTIONAL ALPHA COUNTER

Application

Internal proportional alpha counters are gas-flow counters useful for measuring levels of alpha activity commonly found in environmental samples. The counters are termed "internal" because the sample is placed into the sensitive part of the detector, and counting gas is purged throughout both counting chamber and sample. Because of this procedure, the counter can be contaminated by the sample. At the same time, this essentially eliminates all losses but those from self-absorption. For samples deposited on a planchet, this counter is as sensitive as any alpha counting system, and can be used to count activities ranging from the lowest to the very highest.

To be detected, the alpha particles have only to penetrate the sample itself and produce several ion pairs in the gas. The detector is almost equally sensitive over the inside diameter, although a slight decrease in response is noted at the edge of the counting dish. Any metallic planchet is suitable for counting in an internal proportional counter, but care should be taken to have the solids uniformly distributed in the dish. This is especially true for counting alpha activity if there is much solid material in the sample, as absorption losses from unevenly distributed solids will be inconsistent. Stainless steel dishes are used for most samples, but cannot be used with chlorides, In this case, aluminum must be used.

Operation

The counters are termed "proportional" because they operate at voltages where the size of the pulse is proportional to the number of ion pairs produced. A counting gas mixture of 90 percent argon and 10 percent methane (P-10 gas) is commonly used. The electrons from ion pairs produced in this gas are collected by the central electrode to generate a pulse which is recorded by a scaler. Since alpha particles dissipate more energy in the chamber than beta particles, they produce more ion pairs and consequently large pulses. The smaller beta-gamma pulses, if present, are discriminated against by operating at the relatively low voltage of approximately 1000 V which produces a small amplification. Ability to count only alpha emissions at one voltage and alpha plus beta-gamma emissions at another makes the proportional counter a versatile detector.

If the instrument used for counting samples of high activity is also used for counting those of low activity, background measurements should be more frequent than twice daily to check for contamination. For an unshielded 2 1/4-inch diameter hemisphere and piston, the alpha background is about 0.2 counts per minute.

The alpha counting efficiency for weightless samples is slightly higher than 50 percent. The efficiency for anything but weightless samples is substantially lower due to severe self-absorption losses.

2. ALPHA SCINTILLATION COUNTER - Zinc Sulfide Screen

Application

The alpha scintillation counter is used for counting flaky or dusty samples such as air samples, that could easily contaminate an internal proportional counter. Uneven self-absorption losses from these samples can result in poor reproducibility, even with the scintillation counter. This instrument lends itself quite readily to mobile laboratory or field use because no gas supply is needed.

Since the sample is external to the detector, the counting dish or planchet need not be conducting. It should be shallow enough to enable the sample to be placed close to the zinc sulfide screen to minimize air absorption losses. In this configuration, the alpha scintillation counter is almost as sensitive as the internal proportional counter. Samples deposited on membrane filters provide a nearly ideal geometry because they are flat and usually evenly distributed. Self-absorption, however, can be severe for air samples having a high solids content.

Operation

The counter is composed of a zinc sulfide screen (zinc sulfide deposited on lucite or some other transparent substance), and a photomultiplier tube housed in a light-tight box coupled through a cathode follower into a scaler. The voltage that must be used depends on the photomultiplier tube characteristics. The tube should have a uniform photo-cathode which yields a satisfactory plateau. No additional amplification of the pulses is required, and operating voltages are usually less than 2000 V, enabling use of the simplest scalers.

A 2-inch diameter weightless sample close to a 3 1/2-inch screen has a counting efficiency greater than 40 percent. For the same weightless sample, the efficiency will be about 50 percent in an internal proportional alpha counter. The efficiency obtained is much better than for an end window alpha counter, since the zinc sulfide screen has no window.

The background is about 0.2 cpm for a 3 1/2-inch slope per 100 volts.

3. ALPHA SCINTILLATION COUNTER - Liquid Scintillation

Application

The liquid scintillation counter was designed primarily for counting weak beta emitters such as tritium (^3H) and carbon 14. It can also be used successfully for alpha counting. Its chief advantage is that there can be no self-absorption losses in the sample to cause non-reproducible counting. Sample preparation may require some training in radiochemistry.

The samples are mixed with the scintillator and the mixture is placed in low potassium-content glass or polyethylene vials. The alpha counting efficiency approaches 100 percent, and the minimum sensitivity for radium 226 is about 0.2 picocurie per sample for a 100-minute count.

An alternate method is to place the sample in a small vial coated with zinc sulfide powder. With this procedure, the background is about 0.02 counts per minute, and the minimum sensitivity for radium 226 is approximately 0.02 picocurie per sample for 100-minute counting periods.

The liquid scintillation for alpha counting has not been used extensively by the laboratories; but is described here as an alternate to the more specialized radon emanation technique which follows.

Operation

A typical liquid scintillation counter uses two photomultiplier tubes in coincidence. Noise pulses or originating in the tubes can be discriminated against as they are much smaller than alpha pulses. With the coincidence circuit, pulses must occur simultaneously in each tube to be recorded as a count.

4. RADON COUNTER - Gas Phase Counting

Application

The radon counter is used for the determination of radium 226 by measuring radon 222 and its daughters. The daughters of importance to the determination are the short-lived alpha emitters polonium 218, bismuth 214, and polonium 214. Lead 214 which follows polonium 218 in the decay chain is a beta emitter.

Gas phase scintillation counting yields the maximum sensitivity for radium 226. With appropriate sample preparation, it can be used for almost any sample for activities down to 0.01 picocurie per sample if 1,000-minute background and sample counts are made.

Operation

The radium 226 is stored in a sealed tube until the system has reached secular equilibrium (about 28 days) or at least until an amount of radon 222 sufficient for counting has grown into the radium 226 solution. Then the radon gas is purged into the evacuated scintillation cell by using radon-free gas, such as aged air or nitrogen at a low purge rate.

The cell containing the radon must be stored in a light-tight container for approximately 5 hours to enable the radon daughters to reach equilibrium. At equilibrium, for every radon 222 disintegration three alpha particles will be available for counting. This storage time can vary as long as the degree of equilibrium attained is considered in the calculation of radium 226 activity. The storage time should be long enough to allow any actinon (^{219}Rn) or thoron (^{222}Rn) daughters of radium 223 or radium 224, if present, to decay out. It is usually convenient to store the cell in the same light-tight container which houses the photomultiplier tube. From its photomultiplier tube, which detects the light flashes in the cell, to its scaler, this counter is very similar to the zinc sulfide screen already described. The scintillation cell substitutes for the screen as the detector.

The counter is plateaued and calibrated by preparing a standard solution of radium 226 and processing the standard in the same manner as the samples are processed. This determines the efficiency of the purge and the counting.

Counting efficiency for the radon and daughters is approximately 80 percent in a properly coated cell. The background is usually 0.1 count per minute. Each cell must be checked independently on the phototube used for sample counting. It is important to keep the cell and the tube as a matched unit, as each can affect background and counting efficiency.

b. Beta Activity

Beta particles (Beta Activity) are negatively charged electrons ejected from the nucleus when a neutron decays to a proton and an electron. They are not monoenergetic, but have a continuous distribution of energies from zero up to E_{max}, the maximum value reported for the particular radioisotope. Each radionuclide has a different decay scheme which can be used to help identify the constituents of a sample containing beta emitters. Some beta emitters have many E_{max} values due to the way the atom reached a stable state. The average energy of the beta particles from a given emitter is generally taken to be about one third of the maximum. The energy released by decay partially determines the type of instrument needed.

For each instrument described, there is an operating high voltage plateau which will yield consistent counting results. To determine this voltage range, counts are made with a beta emitter which has approximately the same beta energy as the beta emitter in the samples. Counts are made at 50-volt increments. When the counts from one 50-volt increment to the next are within 1 percent of each other, the plateau has been reached and the operating voltage lies in this plateau. Each instrument varies as to the length of the plateau and the best operating voltage. The operating voltage is influenced by the construction of the counter and for a gas ionization detector, the type of gas used. Additional information will be found in the operating manual supplied with the particular instrument and in several references listed at the end of this Section.

1. GEIGER-MUELLER COUNTER

Application

The Geiger-Mueller (GM) counter is a gas ionization detector operated in the Geiger-Mueller region of response. In this application, the gas ionization chamber is called a GM tube. The cathode of this tube is usually a part of its wall, and the anode is a central wire mounted within the cathode. GM tubes contain the counting gas, which is usually a noble gas mixed with a small percentage of some halogen or organic gas to provide quenching. Typical gas mixtures are argon and ethanol, helium and isobutane, and neon and an organic halide.

The GM counter is relatively inexpensive, can be used with every simple rate meters and scalers, has a very low threshold of sensitivity, and is relatively trouble-free. Its high background, low counting efficiency, and inability to differentiate between two or more types of incident radiation can be disadvantageous in some applications.

The GM detectors are used to count samples containing near-tracer levels of activity. Although all pulses produced in the GM tube are the same size whether they originate from a low or high energy beta particle, qualitative identification of beta emitters in samples of high enough activity can be obtained by placing various thicknesses of aluminum absorbers between the sample and the GM tube. In this manner, the endpoint energy, or E_{max}, of the beta emissions from the source can be determined.

Almost any planchet is suitable for the GM detector, and reproducible results will be obtained as long as the planchet diameter is smaller than the diameter of the GM tube itself. In a 11/2-inch thick lead shield, a 11/2-inch diameter tube will have a background of about 30 counts per minute. If the radioactive material is near the edge of a 2-inch diameter planchet, the counting efficiency is less than if the same activity is deposited near the center. Since the sealing window must have a density thickness of about 2 milligrams per square centimeter, and there is an air path between the sample and window, there is a substantial absorption loss for beta energies less than 400 keV (E_{max}).

Operation

The counting efficiency of this instrument is the lowest of those described here, especially for low-energy beta particles, because of the relatively thick window and poor geometry. Significant dead-time losses can occur if the count rate is high (above 10,000 counts per minute) and will usually be a problem only when the instrument is being calibrated at high levels of activity. If sample count rates are excessive, the sample must be counted at a greater distance from the tube. Normal losses due to dead-time are 0.5 percent per 1,000 counts per minute.

A plateau should be obtained at regular intervals to ensure that the GM detector is functioning properly. The plateau should not have a slope of more than 2 percent per 100 volts, and for a new tube should be about 250 volts long. As the tube ages, the plateau will shorten. The operating voltage should be chosen at the lower third of the plateau to prolong the life of the tube. Each tube has its own voltage requirement, but most operate at voltages of less than 2,000.

Once this operating voltage has been determined, the tube should be checked against a performance standard at least three times each day - in the morning, around noon, and at the close of the working day. Background measurements should be made at least twice a day.

Each Geiger-Mueller counter will be accompanied by an operating manual when purchased. It should be consulted for more specific information on calibration and operation of the counter.

2. INTERNAL PROPORTIONAL BETA COUNTER

Application

The internal proportional counter, previously discussed under a. Alpha Activity - 1. Internal Proportional Alpha Counter, is also useful for measuring gross beta activity, where count rates are apt to be higher than in samples chemically separated to contain a single beta-emitting nuclide. It can be a satisfactory instrument for measuring levels of beta activity as low as 5 picocuries per sample if long counting times are used. Even weak beta emitters can be counted, because the sample is within the counter itself. The detector has a 2-pi geometry, and counting efficiency can be as high as 75 percent because of backscatter from the planchet. The increased backscatter is a function of beta energy and of the atomic number of the planchet material.

When beta activity of a sample containing both alpha and beta emitters is to be measured, care must be taken to subtract the count contributed by the alpha emitters from the total count obtained at the beta plateau. The gamma counting efficiency is very low, ranging from approximately 1 percent for 1.0 MeV gammas to approximately 5 percent for 0.1 MeV gammas, and any contributions from gamma emissions can usually be neglected.

Operation

When the internal proportional counter is operated at approximately 1,800 volts, it becomes a beta counter, although alpha and gamma radiations are also counted. There are therefore two plateaus, one for alpha and one for alpha plus beta-gamma emissions. The efficiency for counting alpha activity at voltages in the beta-gamma plateau is some what higher than that obtained at voltages in the alpha plateau. This ratio must be determined if the alpha count is to be subtracted from the beta-plus-alpha count to obtain the corrected beta activity.

The plateau is determined by counting an extended source (2-inch diameter) having approximately the same beta energy as the samples. The operating voltage should be chosen about midway on the beta-gamma plateau. The plateau should be checked at one-month intervals or whenever a new tank of gas is installed.

The background at the beta plateau of an unshielded chamber of 2 1/4-inch internal diameter is approximately 55 counts per minute and is about 30 counts per minute for one shielded with 2 inches of lead.

The counting efficiency is a function of energy, sample thickness, and backing material of the sample. Distribution of activity in the planchet is relatively unimportant, although the efficiency is somewhat lower at the edges of the dish. Table B shows typical efficiencies for counting radionuclides of various average beta energies in the internal proportional counter.

It is extremely important that the samples be conducting to ground to eliminate a static charge buildup which will produce sporadic and inconsistent counts. This static buildup can be compared to formation of a cloud over the sample. During cloud formation, the counts would be suppressed, and when the cloud is removed (discharged) a burst of counts would be observed. Therefore, a non-metal planchet such as polyethylene or glass cannot be used.

The manufacturer's operating manual should be consulted for additional information on operating characteristics and trouble shooting.

Table B. Typical counting efficiencies of the internal proportional beta counters for weightless samples

Radionuclide	Average β Energy (MeV)	Total Efficiency (%)
^{95}Nb	0.046	52
^{147}Pm	0.062	58
^{131}I	0.180	65
^{36}Cl	0.252	70
^{137}Cs-^{137m}Ba	0.268	70
^{90}Sr-^{90}Y	0.566	73
^{89}Sr	0.589	73
^{91}Y	0.615	73
^{32}P	0.693	73
^{90}Y	0.931	73

Counting chamber diameter: 2 1/4 inches;
Planchet dimensions: 2-inch diameter by 1/4 inch high,
0.018-inch thick stainless steel

3. END-WINDOW PROPORTIONAL COUNTER

Application

End-window proportional counters have application for counting moderate to high activity level and for samples that cannot be fixed well in the planchet or sample holder. Large counters of this type can be used to count high-volume air samples such as those collected on 8-inch by 10-inch glass fiber filters.

The end-window instrument is less sensitive than the internal proportional counter because of poorer sample geometry and because of absorption losses in the air path and window. Although a very thin window can be utilized for alpha counting, it is not particularly recommended because of the end-window detector's poor efficiency for alpha detection. It can be used as a screening instrument for alpha and low-energy beta emitters, but is unsuitable for quantitative measurement of these emitters.

Almost any type of planchet can be used with the end-window counter, but for reproducible results its diameter must be somewhat shorter than that of the window. There is no problem of static charge build-up because the sample is external to the counter.

Operation

The end-window proportional counter operates on essentially the same principle as the internal proportional counter. The detector ordinarily uses P-10 gas (argon-methane mixture).

Counting efficiency of the end-window proportional counter is little more than half that of the internal proportional counter. Again, this is due to absorption of some beta particles in the air path and in the window material. The lower energy betas (E_{max} less than 300 KeV) and the backscattered events are especially attenuated.

Background is lower than that of the internal proportional counter because there is less detector area. Contamination is not as apt to occur because the sample is external to the counter. Background counts need be taken only once or twice daily unless a sample contacts the window or contaminates the inside of the shield or sample holder.

Plateaus should be taken at the same intervals and for the same reasons as for the internal proportional counter. Additional information on operation or trouble shooting will be found in the manufacturer's operating manual.

(4) LOW-BACKGROUND BETA COUNTER

Application

The low-background beta counter is designed for measuring low levels of activity. Its primary application is for the determination of strontium fission products in the environment, and use of the counter should be restricted to samples of low to moderate activity. It has the highest sensitivity of any common laboratory beta counter for samples mounted in counting dishes. The instrument is sensitive to about 0.5 picocurie per sample for energetic beta-emitters counted 100 minutes or more. It is possible to determine activity levels of less than one picocurie per sample for the energetic strontium beta emitters. For betas having E^{max} less than 0.3 MeV, other counters should be considered.

The counting dish can be of almost any material depending on the chemical procedure required for sample preparation. The diameter of the dish should be smaller than the diameter of the counter to obtain reproducible results. The two most commonly used planchets are the stainless steel dishes and the nylon ring and dish types. For some applications, the nylon ring and disk offers the advantages of producing a more uniform sample and is easier to prepare. A thin Mylar cover is placed over the sample to eliminate loss of sample and to prevent contamination of the counting chamber. The absence of a wall on the planchet and the protective cover over the sample allows the samples to be placed closer to the window of the counter, thereby yielding higher counting efficiencies.

Operation

A low-background beta counter is shielded by two inches of lead or equivalent to stop external gamma rays, and by coincidence circuitry to reduce the contribution from cosmic interactions in the shielding material. The circuitry consists of one or more guard detectors placed over the central detector(s) so that any radiation interacting in both the guard and central detector(s) is not recorded as a count.

The detectors are commonly GM end-window tubes with thin windows (less than 0.1 milligram per square centimeter) and a positive gas pressure maintained by a low purge rate. Sometimes P-10 gas (argon-methane mixture) is used, and the counter is then operated as a proportional counter. However, the detector is usually operated in the Geiger-Mueller region, using helium-isobutane as the gas, where the pulses produced are very large and no additional amplification is required. The system is usually packaged as a complete unit of central detector, guard tubes, shielding, and scaler. The scaler will have the required high voltage and separate registers for the central detector(s) and guard tube(s).

The counting efficiency for yttrium 90 distributed uniformly on a 2-inch stainless steel planchet placed close to a 2 1/4-inch detector, is about 42 percent, and is about 50 percent for the nylon ring and disk mount.

For samples having activity levels of less than 5 picocuries per sample, adequate counting times in these instruments are usually 100 minutes or overnight. Background measurements must be made at least once daily for 100 minutes or more. In the Geiger-Mueller mode of operation the background count rate should be about 1 count per minute for a 2-inch diameter tube. When the detector is operated in the proportional mode, the background count rate can be further reduced by use of discriminators.

5. LIQUID SCINTILLATION COUNTER

Application

This instrument is particularly well suited to counting low-energy beta emitters such as tritium or carbon 14. By mixing the sample with the liquid scintillation medium, self-absorption losses are eliminated. Counting efficiency can approach 100 percent for high-energy beta emissions, but for those of carbon 14 and tritium the efficiency is much less. Use of the liquid scintillation counter is not limited to counting low-energy beta emitters, but it or a method equivalent to it, such as gas phase counting, is essential for counting weak beta emitters with reliability.

Early models required placing the phototubes in a freezer to reduce the dark current noise to an acceptable level. Recent improvements, however, make this unnecessary for all but the lowest levels of activity (less than about 30 picocuries per sample for ^{14}C).

Operation

In a liquid scintillation counter, the samples and scintillator form a homogeneous mixture in solvents such as toluene, xylene, and 1,4-dioxane. Light flashes produced in the mixture are detected and amplified by the photomultiplier tube.

The samples are placed into a transparent or translucent bottle to enable the light to be transmitted to the phototubes. As in any other counter, the calibration standard must contain the same radionuclide prepared in the same medium. The background count rate is determined by counting the activity of a bottle containing both solvent and scintillator, since this mixture is the sensor and a slight amount of activity may be present in the material itself. Background counts should be taken at least once each day.

Most of the liquid scintillation counting systems designed for low level counting place two phototubes in coincidence. Only one is considered the detector, and it is connected to the analyzer. When both tubes receive a pulse simultaneously, a count is registered equivalent to the energy of the radiation absorbed. When only one tube sees the pulse, no count is recorded. In this way the dark current, or photomultiplier background, is practically eliminated. A discriminator is used to eliminate additional low energy pulses, but the weaker pulses from soft betas are also eliminated, thereby reducing the counting efficiency for these emissions.

The beta energy of the nuclide to be counted determines the discriminator setting and window width. Window width is also dependent on whether or not there is more than one beta emitter present.

Most liquid scintillation counters are coupled to an analyzer with at least two channels. This enables more than one beta emitter to be analyzed at the same time as long as their E_{max} values differ by at least a factor of three.

More detailed information on operation of the liquid scintillation counter will be found in the manufacturer's instruction manual.

c. Gamma Activity

Gamma rays are electromagnetic radiations emitted by the nucleus as photons of discrete energies. The photons have no mass and have high penetrating power. Therefore, sample preparation is generally much less of a problem for counting gamma emitters than for counting either alpha or beta emitters.

The method for measuring gamma activity is basically to capture part or all of the energy released through decay, and to convert it into a current flow or pulse which can be recorded. The gross count can be taken by totaling the pulses recorded on scalers. For spectral analysis, the pulses are sorted out by energy, and the number of pulses counted in each energy band is stored separately. A pulse height analyzer is used for this spectral analysis.

1. SEMICONDUCTOR DETECTORS

A semiconductor is a material that can act as an insulator or as a conductor. In electronics the term solid state is often used interchangeably with semiconductor, but in the detector field the term can obviously be applied to solid scintillators. Therefore, semiconductor is the preferred term for those detectors which are fabricated from either elemental or compound single crystal materials having a band gap in the range of approximately 1 to 5 eV. The group IV elements Silicon and Germanium are by far the most widely-used semiconductors, although some compound semiconductor materials are finding use in special applications as development work on them continues.

Table C shows some of the key characteristics of various semiconductors as detector materials:

Table C. Element vs. Band Gap

Material	Z	Band Gap	Energy/e-h pair (eV)
Si	14	1.12	3.61
Ge	32	0.74	2.98
CdTe	48-52	1.47	4.43
HgI_2	80-53	2.13	6.5
GaAs	31-33	1.43	5.2

Semiconductor detectors have a P-I-N diode structure in which the intrinsic (I) region is created by depletion of charge carriers when a reverse bias is applied across the diode. When photons interact within the depletion region, charge carriers (holes and electrons) are freed and are swept to their respective collecting electrode by the electric field. The resultant charge is integrated by a charge sensitive preamplifier and converted to a voltage pulse with an amplitude proportional to the original photon energy.

Since the depletion depth is inversely proportional to net electrical impurity concentration, and since counting efficiency is also dependent on the purity of the material, large volumes of very pure material are needed to ensure high counting efficiency for high energy photons.

Prior to the mid-1970's the required purity levels of Si and Ge could be achieved only by counter-doping P-type crystals with the N-type impurity, lithium, in a process known as lithium-ion drifting. Although this process is still widely used in the production of Si(Li) X-ray detectors, it is no longer required for germanium detectors since sufficiently pure crystals have been available since 1976.

The band gap figures in Table C signify the temperature sensitivity of the materials and the practical ways in which these materials can be used as detectors. Just as Ge transistors have much lower maximum operating temperatures than Si devices, so do Ge detectors. As a practical matter both Ge and Si photon detectors must be cooled in order to reduce the thermal charge carrier generation (noise) to an acceptable level. This requirement is quite aside from the lithium precipitation problem which made the old Ge(Li), and to some degree Si(Li) detectors, perishable at room temperature.

The most common medium for detector cooling is liquid nitrogen, however, recent advances in electrical cooling systems have made electrically refrigerated cryostats a viable alternative for many detector applications.

In liquid nitrogen (LN_2) cooled detectors, the detector element (and in some cases preamplifier components), are housed in a clean vacuum chamber which is attached to or inserted in a LN_2 Dewar. The detector is in thermal contact with the liquid nitrogen which cools it to around 77°K or -200°C. At these temperatures, reverse leakage currents are in the range of 10^{9} to 10^{-12} amperes.

In electrically refrigerated detectors, both closed-cycle Freon and helium refrigeration systems have been developed to eliminate the need for liquid nitrogen. Besides the obvious advantage of being able to operate where liquid nitrogen is unavailable or supply is uncertain, refrigerated detectors are ideal for applications requiring long-term unattended operation, or applications such as undersea operation, where it is impractical to vent LN_2 gas from a conventional cryostat to its surrounding.

Detector structure

The first semiconductor photon detectors had a simple planar structure similar to their predecessor, the Silicon Surface Barrier (SSB) detector (see section on particle detection). Soon the grooved planar Si(Li) detector evolved from attempts to reduce leakage currents and thus improve resolution.

The coaxial Ge(Li) detector was developed in order to increase overall detector volume, and thus detection efficiency, while keeping depletion (drift) depths reasonable and minimizing capacitance. Other variations on these structures have come, and some have gone away, but there are several currently in use.

Performance

Resolution: Semiconductor detectors provide greatly improved energy resolution over other types of radiation detectors for many reasons. Fundamentally, the resolution advantage can be attributed to the small amount of energy required to produce a charge carrier and the consequent large "output signal" relative to other detector types for the same incident photon energy. At 3 eV/e-h pair (see Table 1) the number of charge carriers produced in Ge is about one and two orders of magnitude higher than in gas and scintillation detectors respectively. The charge multiplication that takes place in proportional counters and in the electron multipliers associated with scintillation detectors, resulting in large output signals, does nothing to improve the fundamental statistics of charge production.

The resultant energy reduction keV (FWHM) vs. energy for various detector types is illustrated in Table D.

Table D. **Energy Resolution vs. Detector Type**

Energy (keV)	**5.9**	**1.22**	**1.332**
Proportional Counter	1.2	-	-
X-ray NaI(Tl)	3.0	12.0	-
3x3 NaI(Tl)	-	12.0	60
Si(Li)	0.16	-	-
Planar Ge	0.18	0.5	-
Coaxial Ge	-	0.8	1.8

Efficiency: The efficiency of detector materials is closely related to their atomic number. Table C gives the atomic number of common semiconductor detector materials and it is easy to see why silicon is relegated to use in the relatively low energy X-ray range. Although the compound semiconductors have high atomic numbers, charge trapping and other practical considerations have denied us large volume devices which are necessary for high energy gamma-ray spectroscopy. The discussions of efficiency center mainly on the Ge detector.

At low energies, detector efficiency is a function of cross-sectional area and window thickness while at high energies total active detector volume more or less determines counting efficiency. Detectors having thin contacts, e.g. Si(Li), Low-Energy Ge and Reverse Electrode Ge detectors, are usually equipped with a Be cryostat window to take full advantage of their intrinsic energy response.

Coaxial Ge detectors are specified in terms of their relative full-energy peak efficiency compared to that of a 3x3 in. NaI(Tl) Scintillation detector at a detector to source distance of 25 cm. Detectors of greater than 100% relative efficiency have been fabricated from germanium crystals ranging up to about 75 mm in diameter. About 2 kg of germanium is required for such a detector.

2. GROSS GAMMA COUNTER

Application

A gross gamma counter is frequently used as a screening device to provide a rapid means of selecting from a group of environmental samples those which have a relatively high activity. It is also used in tracer experiments where a known radionuclide is to be counted, as in determining chemical yield for an analytical procedure by use of a tracer. Depending on the type of sample to be counted, a solid or a well crystal detector can be used for these applications.

Operation

A relatively small crystal scintillator, such as a 2-inch by 2-inch right cylinder, is frequently used for gross gamma counting because total absorption of the gamma rays is not necessary. Any partial interaction with the crystal which produces a pulse above the threshold will register as a count. Shielding is desirable, but it need not be massive. About 1 inch to 1-1/2 inches of lead, or equivalent, is adequate for most applications. With this amount of shielding, a 2-inch by 2-inch crystal will have a background of approximately 300 counts per minute.

A photomultiplier tube detects the light flashes produced in the crystal scintillator. The output from this tube is coupled to a scaler having an amplifier, or an amplifier is inserted as a separate component. Although a simple scaler may be adequate, low-energy sources, in particular, may require a relatively stable high voltage supply.

The gross gamma counter is the simplest and cheapest of the gamma counters routinely used in the laboratory. Its principal limitations are that it has no energy discrimination and that its relatively high background makes it useful only for samples of fairly high activity.

As for all gamma counting, gross gamma counting requires minimal sample preparation because self-absorption losses are not a major problem. For the same reason, almost any sample configuration can be used. It is important, however, to maintain a constant geometrical configuration for a given sample type and to calibrate with a standard having the same configuration as the sample. Even where only relative counts are needed, as for checking chemical procedures, the geometry must be kept constant for all count rates being compared, or it must be calibrated for a variety of sample sizes and configurations.

For most gamma measurements, a discriminator should be used to eliminate much of the low-energy component of background. An optimum setting can be obtained by taking counts at various discriminator settings, both from background and from a source. The best discriminator setting for a given gamma energy is the setting at which the highest value of the ratio S^2/B is obtained, where S is the sample count rate and B is the background count rate.

Gross gamma counting efficiencies vary with energy, but they are quite high because only a partial interaction is needed. For spectrometry, however, total interaction of the gamma photon is needed if the pulse is to be recorded under the photopeak.

Background should be measured twice daily or more often if high activity sources are being moved about nearby. A change in position of these sources will affect the background.

3. GAMMA SPECTROMETER

Application

Gamma spectrometry is used extensively to identify radionuclides. The spectrometer becomes a quantitative tool with suitable calibration and background data accompanied by measurement of spectral interferences. Self-absorption losses are negligible, so large-volume samples can be scanned with little or no prior preparation.

The spectrometers are pulse height analyzers of the single-channel or multichannel type. They are now coupled to computer systems giving them great flexibility and many "automated" software packages to perform calculations at high speeds. A pulse height analyzer sorts pulses according to energy expended in the detector. The multichannel analyzer simultaneously sorts and records pulses falling in many different energy bands or increments in a given time interval. For a given analyzer, the total energy range scanned is divided into equal increments called channels. The single channel analyzer detects and records pulses in only one energy band at a time. The single-channel analyzer is adequate for counting tracer levels of activity, but for low activity samples, the multichannel analyzer is essential because of the excessive counting time required to cover a spectrum channel by channel with a single-channel analyzer.

Either a 3-inch by 3-inch or 4-inch by 4-inch right cylindrical thallium-activated sodium iodide [NaI(Tl)] crystal provides optimum sensitivity for gamma energies up to 2 MeV. These sizes are therefore used for environmental samples containing fallout debris and natural radioactivity. For low-activity gamma spectrometry, the phototube base should be removed and a mu-metal cover should shield the tube to prevent magnetic fields from distorting the spectra.

Low-energy gamma spectrometry ($E\gamma < 150$ keV) requires a very thin crystal less than 0.5 inches thick enclosed by a thin protective can. This thin crystal can also be used to detect the X rays emitted by some radionuclides to help identify the components of a sample spectrum. Very high-energy gamma photons ($E\gamma >$ 2 MeV) are not seen efficiently by the thin detector. Since the predominant gamma energies of gamma emitters in most environmental samples are less than 2 MeV, large crystals will not be discussed.

The Crystal Detector

A detector for the gamma spectrometer systems in addition to the solid state detectors as in section 1. is a sodium iodide crystal optically coupled to a photomultiplier tube. The tube is connected into the analyzer through a pre-amplifier and linear amplifier. The analyzer records each pulse in its proper channel or energy increment. Each channel represents a certain amount of energy absorbed in the crystal. The spectrum shows a characteristic photopeak at the full energy of the gamma ray, and at lower energy regions shows counts which arise from partial interactions of the gamma rays with the crystal. By knowing the decay schemes of radionuclides, it is possible to identify specific isotopes by their gamma energies.

Crystal size is very important in gamma spectrometry. If too large a crystal for the application is chosen, the background will be unnecessarily high. If a crystal is too small for the energies being scanned, the photopeak counting efficiency will be low because few of the gamma photons will expend their full energy in the crystal. The detector is energy dependent, the efficiency being lower for higher energies. This is especially noticeable in pulse height analysis, because the entire photon energy must be spent in the crystal to be recorded under the photopeak. This type of interaction is called photoelectric, and it is the most important interaction in gamma spectrometry.

All higher energy photons contribute to a lower region of the energy spectrum because of Compton interaction. In the Compton interaction, only a portion of the total energy is absorbed in the crystal and the remainder is scattered out of the crystal. The energy absorbed is recorded as a pulse in a channel below that of the photopeak. The amount of energy lost by this process is somewhat energy dependent, but it can be from almost full energy to only a small percentage of the total energy of the gamma photon. Frequently there are multiple Compton interactions, and if they occur in the crystal, the total energy absorbed will be additive. In some instances, the sum may equal the full gamma energy of the photon, and the pulse will then be recorded under the photopeak.

Another form of gamma interaction important to gamma spectrometry is pair production. This process occurs only at gamma energies above 1.02 MeV. One electron of the pair is negatively charged, and the other positively. The positively charged electron, positron, is slowed down until it interacts with a negatively charged electron and both are annihilated. Their rest mass energy of 1.02 MeV is converted to two photons, each having an energy of 0.51 MeV, which are emitted in opposite directions.

This conversion results in a peak in the gamma spectrum at 0.51 MeV. If the counting geometry is such that both of the photons can interact with the detector simultaneously (as in a well-type detector), a sum peak at 1.02 MeV will be observed. If, however, one or both entirely escape the detector, there will be peaks observed at energies of 0.51 or 1.02 MeV less than the full energy of the photon.

Counting Efficiency

The term "total efficiency" denotes the ratio of the net gamma counts observed in a peak spectral region to the total number of disintegrations of the radionuclide of interest in the same period of time, and is determined by the following equation.

$$\text{Total efficiency (\%)} = \frac{\text{Observed counts in peak region - background in peak region}}{\text{Total disintegrations}} \times 100$$

If the activity of a standard is given in terms of total disintegration rate, the gamma abundance is not needed to determine the photopeak efficiency of a particular radionuclide. A radioactive standard of ruthenium 106-rhodium 106 (^{106}Ru-^{106}Rh) will serve as an example. Let the following information be given:

^{106}Ru activity in the sample = 100,000 dpm
^{106}Rh (daughter of ^{106}Ru) gamma energy = 0.51 MeV
Abundance of the 0.51 MeV gamma of ^{106}Rh = 20%
Net gamma count rate between 0.44 and 0.56 MeV = 2000 cpm

The equation can then be written in one of three ways:

1) $$^{106}\text{Ru or }^{106}\text{Rh Eff. (\%)} = \frac{\text{Net cpm in peak region}}{\text{dpm in sample of }^{106}\text{Ru or }^{106}\text{Rh}} \times 100$$

$$= \frac{2{,}000}{100{,}000} \times 100$$

$$= 2\ \%$$

2) $$^{106}\text{Ru-}^{106}\text{Rh Eff. (\%)} = \frac{\text{Net cpm in peak region}}{\text{dpm in sample of }^{106}\text{Ru-}^{106}\text{Rh}} \times 100$$

$$= \frac{2{,}000}{200{,}000} \times 100$$

$$= 1\ \%$$

3) $$\text{0.51 MeV Eff. (\%)} = \frac{\text{Net cpm in peak region}}{\text{dpm in sample x gamma abundance}} \times 100$$

$$= \frac{2{,}000}{100{,}000 \times 0.20} \times 100$$

$$= 10\%$$

In the third case, the efficiency for the 0.51 MeV gamma photons would actually be less than 10% because the 2,000 counts in the photopeak region include Compton interactions from higher energy gamma emissions from ^{106}Rh.

Graphs of gamma efficiency plotted against gamma energy can be very useful if the gamma abundance values are accurately known and if the standard is precisely rated. If a range of gamma energies is obtained and the gamma abundances are known, it is possible to predict the gamma efficiency for an unknown radionuclide.

Resolution

Resolution is a measure of the sharpness of a photopeak and it determines the ability of a detector to separate or distinguish between the peaks of gamma rays having similar energies. Without good resolution, photopeaks of similar energies are difficult to quantify.

Resolution is calculated by the following formula:

The units of energy can be either channel number of MeV, keV, etc. for purposes of calculation.

$$\text{Resolution (\%)} = \frac{\text{peak width in energy at half maximum count rate}}{\text{peak energy}} \times 100$$

Resolution is a function of energy, and in general it decreases with increasing energy. Cesium 137 has been generally accepted as the standard for comparison of resolution. It is therefore important to specify the energy in reporting resolution when it has been determined at photopeaks other than 0.662 MeV peak of cesium 137. A typical resolution for a 4-inch by 4-inch NaI(T1) crystal with a 3-inch phototube is about 8.5 percent of cesium 137.

Linearity

Before any calibration or sample counting can be attempted, the analyzer must be linear. To determine this, a combination of accurately known gamma energies covering a substantial portion of the spectral region of interest must be measured. Gamma energy is then plotted against the channel number at which its photopeak occurs. Zero energy should be at the origin, and a nearly straight line with a slope approximately equal to one should be obtained. If the analyzer is linear, channel number 25 should represent one-fourth the energy represented by channel number 100.

Non-linearity can be corrected by analyzer adjustments described in the manufacturer's instruction manual. Daily checks should be made to ensure that the system remains linear.

Procedures for Interpreting Gamma Spectra

The advent of multichannel pulse height analysis gamma spectrometry initiated an entirely new method of collecting data. At first, all results were qualitative, but invariably the question arose of how much of each was present. The questions was first asked in reference to iodine 131 in milk samples because of the significance of this radionuclide as a potential health hazard.

The first approach to quantitating individual components in a mixture of gamma emitters was to smooth off the bottom of each peak at the Compton continuum, and relate area under that peak, in counts per minute, to the disintegration rate of the nuclide represented. As a refinement of this, spectrum stripping was developed. In this procedure, individual components were graphically separated. Next came the simultaneous equation approach for unscrambling the spectrum, and recently the least squares method and modifications of it have been used.

Before any spectral analysis can be performed, a library of spectra must be obtained for all the radionuclides of interest in all the various sample sizes and shapes. This channel-by-channel spectral information is necessary not only for calibration purposes but also for quantifying the data obtained from radionuclide mixtures.

Compton Continuum Subtraction

The easiest way to obtain an estimate of the activity for each radionuclide in a spectrum is to smooth off each photopeak and determine the contribution to that photopeak from the gamma photon occurring at that energy.

The difference between the count rate on the dotted line and the count rate at the peak represents the contribution from the gamma photon producing the peak. As an example, channel 18 has 5000 counts and the dotted line passes through the 1000-count portion of the graph. The contribution of this photopeak to channel 18 is 4000 counts. The 1000 counts represents the Compton contribution from gamma photons of higher energy. If the counting efficiency for this radionuclide was determined over five channels, then two channels on each side of the peak must be used to obtain a corrected count rate. The net counts per minute are then divided by the efficiency to obtain the disintegration rate.

This method is satisfactory when the peak is fairly well defined and when no other photopeaks are immediately adjacent and overlapping the peak being quantified. The method only corrects for Compton interference, gamma photons interacting with less than their full energy. At best, the corrected result is an approximation which is quite satisfactory if the photopeak is relatively high compared to the interference. It is not satisfactory, however, if the peak is only slightly above the Compton continuum. The placement of the dotted line involves some subjective judgment, but this is not a serious problem when the same analyst interprets spectra from both standard and sample.

D. METHODS OF ANALYSIS

(1) Gamma-emitting Radionuclides

(a) Iodine-131, Barium-140, and Cesium-137 in Milk and Other Foods

Gamma-Ray Spectroscopic Method

A. Principle

Applicable to ^{131}I, ^{140}Ba, and ^{137}Cs in fluid milk preserved with HCHO and I and Cs in foods. Because of the nature of gamma-emitting radionuclides, attenuation of gamma-rays in food slurries or mixture would be similar to that of milk or H_2O. Unlike in milk samples, other radionuclides might be present in foods. Therefore, before performing calculation, gamma-ray energy spectrum should be inspected for any radionuclides besides ^{40}K, ^{131}I, ^{137}Cs, and ^{140}Ba. Since cessation of above-ground weapons testing in the 1960s, no other gamma-ray emitters have been regularly observed or detected in food. (Should any be detected, matrix technique should be expanded using standard source for suspected radionuclide to determine matrix coefficient.)

Known volume is placed in counting vessel positioned over and around right cylinder scintillation crystal detector, NaI(Tl), of multichannel gamma spectrometer. Gamma radiation is counted for given time. Accumulated pulses from selected photon energy range are separated from other gamma-emitting radionuclides and background radiation by simultaneous equations. ^{40}K is always present as natural contaminant in food and will contribute counts in 1 or more of photopeak ranges. Mutual interferences among these 4 photopeaks are eliminated by applying matrix technique to separate activities of the 4 nuclides. Measurement of one standard source of each nuclide provides the matrix coefficients.

In special cases, newly formed fission products may be present, e.g., ^{133}I and ^{135}I, which may interfere either through direct overlapping of photopeaks or by contributing Compton-continuum counts. Such interference may be minimized by waiting for decay of short-lived radionuclides, by additional counting following decay, or by chemical separation.

B. Apparatus

(a) *Alignment sources.*—Gamma ray energies, at least 1 near ^{137}Cs spectrum, with well known energies and abundance of gamma rays in photopeaks, for alignment. Solid sources, ca 0.1 μCi, are preferred over liquid sources. ^{207}Bi is satisfactory single source with several photopeaks; ^{137}Cs and ^{60}Co are good pair.

(b) *Counter.*—Low level gamma spectrometer consisting of shielded Tl-activated NaI scintillation detector, 4 × 4 in., coupled to multichannel pulse-height analyzer and readout system.

(c) *Counting vessel* (*Marinelli beaker*).—Use 3.5 L beaker, for 4 × 4 in. detector. Beaker and lid available from plastic laboratory-ware suppliers such as Bel-Art Products, Industrial Rd, Pequannock, NJ 07440-1992, No. H26852 for beaker and No. H26587 for lid.

C. Reagents

(a) *Carrier solutions.*—10 mg/mL. Prepare solutions of CsCl (1.267 g/100 mL), NaI (1.181 g/100 mL), and $BaCl_2 \cdot 2H_2O$ (1.779 g/100 mL). Store in polyethylene or glass bottles.

(b) *Stock standard solutions.*—10 000 pCi/mL. Dilute calibrated solutions of ^{131}I, ^{140}Ba, and ^{137}Cs to approximate indicated strength.

(c) *Potassium-40 stock standard solution.*—1.89 dpm (disintegrations/min) ^{40}K/mg K. Dissolve 240 g KCl (equivalent to 126 g K) in 3 L H_2O in Marinelli beaker and dilute to 3.5 L.

(d) *Calibrating solutions.*—For ^{137}Cs and ^{140}Ba, add 3-5 mL carrier solution, (a), to 3 L H_2O in Marinelli beaker, mix, add convenient amount of stock standard solution (b), sufficient to reduce counting error to ca 1% when counted within 10–100 min, mix, adjust pH to 3.5–4.5, and dilute to 3.5 L. Prepare ^{131}I solution similarly, but adjust pH to 8.5.

D. Preparation of Sample

No special preparation is needed for milk samples. For other foods, do not include inedible material such as bone, apple cores, nut shells, and egg shells as part of sample. Homogenize sample in blender or mechanical homogenizer. Dietary samples prepared for consumption do not require blending, but must be sufficiently mixed to ensure representative sample.

E. Determination

Using alignment sources centered on detector, adjust spectrometer to cover range at least between 0 and 2 MeV, in intervals (channels) of 10 or 20 keV. Adjust voltage or gain control so that the 2 gamma photopeaks of standard fall in their appropriate channels. Check and adjust alignment daily.

Place Marinelli beaker containing 3.5 L calibrating solution (d), over detector, and count standard for time (10–100 min) sufficient to reduce counting error to ca 1%. Repeat with each calibrating solution and with H_2O. Recalibrate spectrometer yearly or more frequently if gamma ray resolution changes.

Transfer 3.5 L well mixed milk sample at room temperature into Marinelli beaker, place over detector, and count 100 min or time sufficient to give desired counting statistics.

F. Calculations

(a) *Counter efficiency.*—Total individual counts observed in channels of photopeak range for each calibrating solution. Subtract total background count for same photopeak range. Divide net count by counting time in min and amount of radionuclide in pCi, and record cpm/pCi for each.

(b) *Interference coefficients.*—When counting standard solution of each radionuclide, ^{131}I, ^{137}Cs, ^{140}Ba, ^{40}K, ratio of net counting rate in energy range of each of the other radionuclides to net counting rate in its own photon energy range gives its fractional interfering coefficient for each of the other energy ranges, e.g., ^{131}I ratio of net counting rate in ^{137}Cs energy range to net counting rate in ^{131}I photon energy range gives its fractional interfering coefficient for ^{137}Cs energy range.

Designate counting rate for ^{131}I, ^{140}Ba, ^{137}Cs, and ^{40}K with symbols I, B, C, and K, respectively. Designate net counting rates (observed—background) in their respective photon energy ranges as N_i, N_b, N_c, and N_k, respectively. Then, f, fractional coefficients or contributions of nuclide in particular range, is designated by 2 lower case subscripts; first one indicates nuclide contributing counts to energy range (column) and second, nuclide photon energy range (row). The following 4 equations:

$$\begin{array}{ll} (1) & N_i = I + f_{bi} B + f_{ci} C + f_{ki} K \\ (2) & N_b = f_{ib} I + B + f_{cb} C + f_{kb} K \\ (3) & N_c = f_{ic} I + f_{bc} B + C + f_{kc} K \\ (4) & N_k = f_{ik} I + f_{bk} B + f_{ck} C + K \end{array}$$

can be solved simultaneously by matrix algebra, using inversions to provide numerical constants W, X, Y, and Z in equations 5, 6, 7, and 8. These constants are used to solve for concentration of each of these 4 nuclides in sample. Net counting rate for each nuclide is:

$$\begin{array}{lll} (5) & ^{131}\mathrm{I} & = I = W_1N_i + W_2N_b + W_3N_c + W_4N_k \\ (6) & ^{140}\mathrm{Ba} & = B = X_1N_i + X_2N_b + X_3N_c + X_4N_k \\ (7) & ^{137}\mathrm{Cs} & = C = Y_1N_i + Y_2N_b + Y_3N_c + Y_4N_k \\ (8) & ^{40}\mathrm{K} & = K = Z_1N_i + Z_2N_b + Z_3N_c + Z_4N_k \end{array}$$

Calibration to derive values for constants in equations 5, 6, 7, and 8 is applicable as long as instrument alignment and mode of operation remain constant and gamma-emitting nuclides are limited to the 4 elements in matrix. Long-hand inversion of 4 × 4 matrix is tedious and subject to mistakes. Use of computer is recommended to provide numerical constants for equations 5–8. Thereafter, desk calculations can determine concentrations of ^{131}I, ^{140}Ba, ^{137}Cs, and ^{40}K in samples in absence of computer by summing counts in each photopeak, subtracting background, and applying equations 5–8.

(c) *Iodine-131, barium-140, cesium-137, potassium-40 activities.*—From spectral gamma counts of sample, substitute net value from equations 5 through 8 and convert net counts/min for each nuclide to pCi/L milk at time of counting:

$$\begin{array}{ll} ^{131}\mathrm{I(pCi/L)} & = (\text{net cpm})_i/(E_i \times V) \\ ^{140}\mathrm{Ba(pCi/L)} & = (\text{net cpm})_b/(E_b \times V) \\ ^{137}\mathrm{Cs(pCi/L)} & = (\text{net cpm})_c/(E_c \times V) \\ ^{40}\mathrm{K(pCi/L)} & = (\text{net cpm})_k/(E_k \times V), \end{array}$$

where E_i, E_b, E_c, E_k = counting efficiency/pCi from standard solutions for ^{131}I, ^{140}Ba, ^{137}Cs, ^{40}K, respectively, and V = sample volume, L.

(b) Iodine-131, Ruthenium-106, and Cesium-137 in Foods

Gamma-Ray Spectroscopic Method

A. Principle

Applicable to ^{131}I, ^{106}Ru, and ^{137}Cs in foods. Because of the nature of gamma-emitting radionuclides, attenuation of gamma-rays in food slurries or mixture would be similar to that of milk or H_2O. Unlike in milk samples, other radionuclides might be present in foods. Therefore, before performing calculation, gamma-ray energy spectrum should be inspected for any radionuclides besides ^{40}K, ^{131}I, ^{137}Cs, and ^{106}Ru. Since cessation of above-ground weapons testing in the 1960s, no other gamma-ray emitters have been regularly observed or detected in food except ^{134}Cs. (Should any ^{134}Cs be detected, matrix technique should be expanded using standard source for suspected radionuclide to determine matrix coefficient.)

Known volume is placed in counting vessel positioned over and around right cylinder scintillation crystal detector, NaI(Tl), of multichannel gamma spectrometer. Gamma radiation is counted for given time. Accumulated pulses from selected photon energy range are separated from other gamma-emitting radionuclides and background radiation by simultaneous equations. ^{40}K is always present as natural contaminant and may contribute counts in one or more of photopeak ranges. Mutual interferences among these 4 photopeaks are eliminated by applying matrix technique to separate activities of the 4 nuclides. Measurement of one standard source of each nuclide provides the matrix coefficients.

In special cases, newly formed fission products may be present, e.g., ^{133}I and ^{135}I, which may interfere either through direct overlapping of photopeaks or by contributing Compton-continuum counts. Such interference may be minimized by waiting for decay of short-lived radionuclides, by additional counting following decay, or by chemical separation.

B. Apparatus

(a) *Alignment sources.*—Gamma ray energies, at least 1 near ^{137}Cs spectrum, with well known energies and abundance of gamma rays in photopeaks, for alignment. Solid sources, ca 0.1 μCi, are preferred over liquid sources. ^{207}Bi is satisfactory single source with several photopeaks; ^{137}Cs and ^{60}Co are good pair.

(b) *Counter.*—Low level gamma spectrometer consisting of shielded Tl-activated NaI scintillation detector, 4 × 4 in., coupled to multichannel pulse-height analyzer and readout system.

(c) *Counting vessel (Marinelli beaker).*—Use 3.5 L beaker, for 4 × 4 in. detector. Beaker and lid available from plastic laboratory-ware suppliers such as Bel-Art Products, Industrial Rd, Pequannock, NJ 07440-1992, No. H26852 for beaker and No. H26587 for lid.

C. Reagents

(a) *Carrier solutions.*—10 mg/mL. Prepare solutions of CsCl (1.267 g/100 mL), NaI (1.181 g/100 mL), and $RuCl_3$ (2.06 g/100 mL). Store in polyethylene or glass bottles.

(b) *Stock standard solutions.*—10 000 pCi/mL. Dilute calibrated solutions of ^{131}I, ^{140}Ba, and ^{137}Cs to approximate indicated strength.

(c) *Potassium-40 stock standard solution.*—1.89 dpm (disintegrations/min) ^{40}K/mg K. Dissolve 240 g KCl (equivalent to 126 g K) in 3 L H_2O in Marinelli beaker and dilute to 3.5 L.

(d) *Calibrating solutions.*—For ^{137}Cs and ^{106}Ru, add 3–5 mL carrier solution, (a), to 3 L H_2O in Marinelli beaker or other standardized volume, mix, add convenient amount of stock standard solution, **(b)**, sufficient to reduce counting error to ca 1% when counted within 10–100 min, mix, adjust pH to 3.5–4.5, and dilute to 3.5 L or other standard volume. Prepare ^{131}I solution similarly, but adjust pH to 8.5.

D. Preparation of Sample

Food samples do not include inedible material such as bone, apple cores, nut shells, and egg shells as part of sample. Homogenize sample in blender or mechanical homogenizer. Dietary samples prepared for consumption do not require blending, but must be sufficiently mixed to ensure representative sample.

E. Determination

Using alignment sources centered on detector, adjust spectrometer to cover range at least between 0 and 2 MeV, in intervals (channels) of 10 or 20 KeV. Adjust voltage or gain control so that the 2 gamma photopeaks of standard fall in their appropriate channels. Check and adjust alignment daily.

Place Marinelli beaker containing 3.5 L calibrating solution or other standardized container, ie 400 mL plastic container (cottage cheese type)(d), over detector, and count standard for time (10–100 min) sufficient to reduce counting error to ca 1%. Repeat with each calibrating solution and with H_2O. Recalibrate spectrometer yearly or more frequently if gamma ray resolution changes.

Transfer 3.5 L well mixed sample at room temperature into Marinelli beaker or other standard volume, place over detector, and count 100 min or time sufficient to give desired counting statistics.

F. Calculations

(a) *Counter efficiency.*—Total individual counts observed in channels of photopeak range for each calibrating solution. Subtract total background count for same photopeak range. Divide net count by counting time in min and amount of radionuclide in pCi, and record cpm/pCi for each.

(b) *Interference coefficients.*—When counting standard solution of each radionuclide, ^{131}I, ^{137}Cs, ^{106}Ru, ^{40}K, ratio of net counting rate in energy range of each of the other radionuclides to net counting rate in its own photon energy range gives its fractional interfering coefficient for each of the other energy ranges, e.g., ^{131}I ratio of net counting rate in ^{137}Cs energy range to net counting rate in ^{131}I photon energy range gives its fractional interfering coefficient for ^{137}Cs energy range.

Designate counting rate for ^{131}I, ^{106}Ru, ^{137}Cs, and ^{40}K with symbols I, R, C, and K, respectively. Designate net counting rates (observed—background) in their respective photon energy ranges as N_i, N_r, N_c, and N_k, respectively. Then, f, fractional coefficients or contributions of nuclide in particular range, is designated by 2 lower case subscripts; first one indicates nuclide contributing counts to energy range (column) and second, nuclide photon energy range (row). The following 4 equations:

$$N_i = I + f_{bi} R + f_{ci} C + f_{ki} K \tag{1}$$
$$N_r = f_r I + R + f_{cr} C + f_{kr} K \tag{2}$$
$$N_c = f_{ic} I + f_{bc} R + C + f_{kc} K \tag{3}$$
$$N_k = f_{ik} I + f_{rk} R + f_{ck} C + K \tag{4}$$

can be solved simultaneously by matrix algebra, using inversions to provide numerical constants W, X, Y, and Z in equations 5, 6, 7, and 8. These constants are used to solve for concentration of each of these 4 nuclides in sample. Net counting rate for each nuclide is:

$$^{131}\mathrm{I} \quad = I = W_1N_i + W_2N_r + W_3N_c + W_4N_k \tag{5}$$
$$^{106}\mathrm{Ru} \quad = R = X_1N_i + X_2N_r + X_3N_c + X_4N_k \tag{6}$$
$$^{137}\mathrm{Cs} \quad = C = Y_1N_i + Y_2N_r + Y_3N_c + Y_4N_k \tag{7}$$
$$^{40}\mathrm{K} \quad = K = Z_1N_i + Z_2N_r + Z_3N_c + Z_4N_k \tag{8}$$

Calibration to derive values for constants in equations 5, 6, 7, and 8 is applicable as long as instrument alignment and mode of operation remain constant and gamma-emitting nuclides are limited to the 4 elements in matrix. Long-hand inversion of 4 × 4 matrix is tedious and subject to mistakes. Use of computer is recommended to provide numerical constants for equations 5–8. Thereafter, desk calculations can determine concentrations of ^{131}I, ^{106}Ru, ^{137}Cs, and ^{40}K in samples in absence of computer by summing counts in each photopeak, subtracting background, and applying equations 5–8.

(c) *Iodine-131, ruthenium-106, cesium-137, potassium-40 activities.*—From spectral gamma counts of sample, substitute net value from equations 5 through 8 and convert net counts/min for each nuclide to pCi/L at time of counting:

$$\begin{aligned}
{}^{131}\mathrm{I(pCi/L)} &= (\mathrm{net\ cpm})_i/(E_i \times V)\\
{}^{106}\mathrm{Ru(pCi/L)} &= (\mathrm{net\ cpm})_b/(E_b \times V)\\
{}^{137}\mathrm{Cs(pCi/L)} &= (\mathrm{net\ cpm})_c/(E_c \times V)\\
{}^{40}\mathrm{K(pCi/L)} &= (\mathrm{net\ cpm})_k/(E_k \times V),
\end{aligned}$$

where E_i, E_r, E_c, E_k = counting efficiency/pCi from standard solutions for ^{131}I, ^{106}Ru, ^{137}Cs, ^{40}K, respectively, and V = sample volume, L.

(c) Cesium-134/137 in Foods

Gamma-Ray Spectroscopic Method

A. Principle

See (b) "Iodine-131, Ruthenium-106 and Cesium-137 in Foods"

B. Apparatus

See as in "A. Principle"

C. Reagents

(a) *Carrier solutions.*—10 mg/mL. Prepare solutions of CsCl (1.267 g/100 mL). Store in polyethylene or glass bottles.

(b) *Stock standard solutions.*—10 000 pCi/mL. Dilute calibrated solutions of ^{134}Cs and ^{137}Cs to approximate indicated strength.

(c) *Potassium-40 stock standard solution.*—1.89 dpm (disintegrations/min) ^{40}K/mg K. Dissolve 240 g KCl (equivalent to 126 g K) in 3 L H_2O in Marinelli beaker and dilute to 3.5 L, or other standard volume.

(d) *Calibrating solutions.*—For ^{137}Cs and ^{134}Cs, add 3-5 mL carrier solution, (a), to 3 L H_2O in Marinelli beaker or other standard container, mix, add convenient amount of stock standard solution, (b), sufficient to reduce counting error to ca 1% when counted within 10–100 min, mix, adjust pH to 3.5–4.5, and dilute to 3.5 L or other standardized volume.

D. Preparation of Sample

Food samples do not include inedible material such as bone, apple cores, nut shells, and egg shells as part of sample. Homogenize sample in blender or mechanical homogenizer. Dietary samples prepared for consumption do not require blending, but must be sufficiently mixed to ensure representative sample.

E. Determination

Using alignment sources centered on detector, adjust spectrometer to cover range at least between 0 and 2 meV, in intervals (channels) of 10 or 20 keV. Adjust voltage or gain control so that the 2 gamma photopeaks of standard fall in their appropriate channels. Check and adjust alignment daily.

Place calibrating solution(d), over detector, and count standard for time (10–100 min) sufficient to reduce counting error to ca 1%. Repeat with each calibrating solution and with H_2O. Recalibrate spectrometer yearly or more frequently if gamma ray resolution changes.

Transfer well mixed sample at room temperature into standardized container, place over detector, and count 100 min or time sufficient to give desired counting statistics.

F. Calculations

(a) *Counter efficiency.*—Total individual counts observed in channels of photopeak range for each calibrating solution. Subtract total background count for same photopeak range. Divide net count by counting time in min and amount of radionuclide in pCi, and record cpm/pCi for each.

(b) *Interference coefficients.*—When counting standard solution of each radionuclide, ^{134}Cs, ^{137}Cs, ^{40}K, ratio of net counting rate in energy range of each of the other radionuclides to net counting rate in its own photon energy range gives its fractional interfering coefficient for each of the other energy ranges, e.g., ^{134}Cs ratio of net counting rate in ^{137}Cs energy range to net counting rate in ^{134}Cs photon energy range gives its fractional interfering coefficient for ^{137}Cs energy range.

Designate counting rate for ^{134}Cs, ^{137}Cs, and ^{40}K with symbols B, C, and K, respectively. Designate net counting rates (observed—background) in their respective photon energy ranges as N_b, N_c, and N_k, respectively. Then, f, fractional coefficients or contributions of nuclide in particular range, is designated by 2 lower case subscripts; first one indicates nuclide contributing counts to energy range (column) and second, nuclide photon energy range (row). The following 3 equations:

(1) $N_b = B + f_{cb} C + f_{kb} K$
(2) $N_c = f_{bc} B + C + f_{kc} K$
(3) $N_k = f_{bk} B + f_{ck} C + K$

can be solved simultaneously by matrix algebra, using inversions to provide numerical constants X, Y, and Z in equations 4, 5, and 6. These constants are used to solve for concentration of each of these 3 nuclides in sample. Net counting rate for each nuclide is:

(4) $^{134}\text{Cs} = B = X_2N_b + X_3N_c + X_4N_k$
(5) $^{137}\text{Cs} = C = Y_2N_b + Y_3N_c + Y_4N_k$
(6) $^{40}\text{K} = K = Z_2N_b + Z_3N_c + Z_4N_k$

Calibration to derive values for constants in equations 4, 5, and 6 is applicable as long as instrument alignment and mode of operation remain constant and gamma-emitting nuclides are limited to the 3 elements in matrix. Long-hand inversion of 3 × 3 matrix is tedious and subject to mistakes. Use of computer is recommended to provide numerical constants for equations 4-6. Thereafter, desk calculations can determine concentrations of ^{134}Cs, ^{137}Cs, and ^{40}K in samples in absence of computer by summing counts in each photopeak, subtracting background, and applying equations 4-6.

(c) *Cesium-134, cesium-137, potassium-40 activities.*—From spectral gamma counts of sample, substitute net value from equations 4 through 6 and convert net counts/min for each nuclide to pCi/L food at time of counting:

$$^{134}\text{Cs(pCi/L)} = (\text{net cpm})_b/(E_b \times V)$$
$$^{137}\text{Cs(pCi/L)} = (\text{net cpm})_c/(E_c \times V)$$
$$^{40}\text{K(pCi/L)} = (\text{net cpm})_k/(E_k \times V),$$

where E_b, E_c, E_k = counting efficiency/pCi from standard solution for ^{134}Cs, ^{137}Cs,
^{40}K, respectively, and V = sample volume, L.

References: JAOAC **56,** 204(1973); **65,** 1039(1982); **69,** 270, 301(1986); **72,** 667(1989).
OFFICIAL METHODS of ANALYSIS, AOAC, 15th EDITION, ARLINGTON 1990, Method 973.67

(2) Beta-emitting Radionuclides

(a) Tritium in Water

Scintillation Spectrometric Method

A. Principle

Sample is distilled to remove quenching materials and non-volatile radioactive materials. Distillation is to dryness to ensure complete transfer of ^3H and background samples are prepared and counted alternately to nullify errors produced by aging of scintillation medium or instrument drift.

B. Apparatus

(a) *Liquid scintillation spectrometer.*—Coincidence-type. Available from Packard Instrument Co., 2200 Warrenville Rd, Downers Grove, IL 60515; and others.

(b) *Liquid scintillation vial.*—20 Ml; low-K glass, polyethylene, nylon, or equivalent bottles, available from manufacturers under (a).

C. Reagents

(a) *Scintillation solution.*—Thoroughly mix 4 g PPO (2,5-diphenyloxazole), 0.05 g POPOP (1,4-bis(5-phenyloxazol-2-yl) benzene), and 120 g solid naphthalene in 1 L spectral grade 1,4-dioxane. (Available from Aldrich Chemical Co., Inc.). Store in dark bottles. Solution is stable 2 months.

(b) *Tritium standard solution.*—Pipet 4 Ml H_2O of known ^3H activity and 16 Ml scintillation solution into scintillation vial, tightly cover vial with screw cap, and mix thoroughly by shaking.

(c) *Background solution.*—Mix 4 mL distilled H_2O (free of ^3H activity to be measured in samples) with 16 mL scintillation solution as in (b).

D. Preparation of Sample

Distill 20–30 mL sample to dryness. Mix 4 mL sample distillate with 16 mL scintillation solution as in C(b).

E. Determination

Prior to counting, dark-adapt and cool sample, background, and standard solutions ca 3 h in instrument freezer at $>2°$ (to prevent solidification of solution with time), or at ambient temperature if ambient temperature liquid scintillation spectrometer is used. Count solutions for total of 200,000 counts or 100 min, whichever is sooner.

F. Calculation

Counting efficiency, $E = (S - B)/D$

^3H, pCi (picoCuries)/mL $= (C - B)/(E \times 4 \times 2.22)$

where S = gross cpm (counts/min) of standard, B = cpm background, D = dpm (disintegrations/min) of ^3H activity in standard, and C = gross cpm for sample.

References: JAOAC 52, 90(1969).
OFFICIAL METHODS of ANALYSIS, AOAC, 15th EDITION, ARLINGTON 1990, Method 969.48

(b) Dehydration of Food Samples for Assay of Tritium in Free Water

INTRODUCTION

This procedure is for a low-temperature, distillation apparatus for Dehydration of Food Samples for Assay of Tritium in Free Water.

REAGENTS

Dry ice or dry ice-alcohol slurry

Ultima Gold Rx (Packard Instruments) or Equivalent

EQUIPMENT

Liquid scintillation spectrometer (Packard Instruments Company, Model 3385) or Equivalent

Polyethylene vials, stored in darkness.

NESLAB RTE 220 Cooling Bath Unit or equivalent

The distillation unit consists of two identical glass tubes joined to a one-piece adapter by means of 35/25 O-ring joints to form a closed distillation system. One tube holds the sample and the second serves as the water trap.

The water trap is cooled in dry ice or in a slurry of dry ice-alcohol contained in a 16 in. x 12 in. x 16 in. high styrofoam box with two-inch-thick walls and bottom. The two-inch thick walls (four in all) and bottom of the box are made by gluing one-inc sheets of styrofoam with "Sticky Styroglue" from Styroformics Division, PolymerTempera, Inc., Medford, Massachusetts, 02155.

Each assembled distillation unit is placed so the adaptor spans the top edge of the styrofoam box. The water trap rest in the cold bath while the sample tube rests in a cylindrical, ceramic heater outside the box. Three distillation units fit on each of two opposite sides. The heaters are supported on small jacks. A one-inch thick sheet of styrofoam is supported over the refrigerant and helps to contain the cold space. The water traps pass through holes in this sheet.

A stopcock serves to direct the distilled water from the sample tube to the water trap and also permits evacuation of the unit via the water trap at the start of distillation. The evacuation is accomplished by connection of the unit to the mechanical vacuum pump through the 18/9 O-ring joint.

Operation of the Distillation Unit

The 3-way stopcock serves to direct the distilled water from the sample tube to the water trap and also permits evacuation of the unit via the water trap at the start of distillation. The evacuation is accomplished by connection of the unit to the mechanical vacuum pump through the 18/9 O-ring joint.

At the start the stopcock is oriented so as to connect the trap to the vacuum pump and the trap is evacuated. The stopcock is then turned to connect the sample tube to the water trap and allow air in the sample tube to pass to the evacuated trap. After a few seconds the stopcock is turned again to connect the water to the vacuum pump and the trap is again pumped out. This succession of operations is repeated until the pressure in the glass unit reads zero on a Dubrovin gauge, 0 - 20 mm range. The stopcock is finally set to allow moisture to distill from the sample tube to the water trap. The distillation should be done with only very gently heating in order that the distillate will be a clear liquid suitable without further treatment for liquid scintillation counting of its tritium content.

DISTILLATION PROCEDURE

1. Orient the 3-way stopcock so the trap is connected to the vacuum pump.

2. Evacuate the trap.

3. Turn the stopcock to connect the sample tube to the water trap and allow air in the sample tube to pass to the evacuated trap.

4. After a few seconds turn the stopcock to connect the water to the vacuum pump and evacuate the trap again.

5. Repeat until the pressure in the glass unit reads zero on a Dubrovin gauge, 0 - 20 mm range.

6. Set the stopcock to allow moisture to distill from the sample tube to the water trap.

7. Distillate with very gently heating.

8. When distillation is complete, remove the sample tube and water trap and seal with their respective closures.

9. Cool the sample tube and trap to room temperature.

10. Wipe off the lubricating grease on the glass joint of the water trap.

11. Wipe off any moisture condensed on the outside of the water trap.

12. Place the trap and sample tube in light-weight, polyurethane containers.

13. Weigh the sample tube and water trap.

14. Record the weight of the dried residue and distillate. Verify proper agreement with the original sample weight and products.

15. Clean the equipment between samples by soaking in boiling detergent solution. This should dislodge or soften the dried residue in the sample tube and remove grease (Apiezon M) from the lubricated joints. If highly radioactive samples are processed, steam the apparatus to remove adsorbed tritium.

COUNTING PROCEDURE

1. Prepare the counting mixture under amber or red light:

    a. Transfer 10 mL of the distillate to a screw-cap, polyethylene liquid scintillation vial.

    b. Add 15 mL of phosphorus solution (scintillation cocktail).

    c. Cap the vial and shake for at least 30 seconds.

    d. Age in a liquid scintillation spectrometer, as specified by manufacturer of LS counter.

4. Count the samples in a liquid scintillation spectrometer. Sum the results of two, 50-minute counts and treat as a single, 100-minute count.

5. Determine the background with a counting mixture containing 10ml of distilled, deep-well water.

6. Determine the counting efficiency with an NBS tritiated water standard.

7. Correct the sample count for background and efficiency.

8. The spectrometer readout provides the results as counts per minute (cpm) per 10 mL. Convert to nanocuries per kilogram (nCi/kg) of food using Equation 1.

$$\mathrm{nCi/kg} = \frac{\mathrm{C - bg \times R}}{\mathrm{W \times E \times V \times 2.22}} \qquad (1)$$

where:

C = count rate in cpm

V = volume of distillate counted = 10 mL
bg = background count rate
R = volume of distillate recovered
W = wgt. of food which produced R

2.22 = conversion factor for dpm to nCi
E = counting efficiency

9. Calculate the counting error using Equation 2.

$$2-\sigma_{ce}=\frac{\pm 2R\times\sqrt{\left(\frac{G}{t}\right)+s^{2}(B)}}{E\times V\times W\times 2.2} \tag{2}$$

where:

G = gross count (sample + background)
$s^2(B)$ = variance of background values
t = time (minutes)
ce = counting error

REFERENCES

1. Stewart, M. L., et al. A Tritiated Water Recovery System. Int. J. of Applied Rad and Iotopes, Volume 23, 1972, Pages 387-8

2. Liebermann and Moghissi in International Journal of Applied Radiation and Isotopes, 21, 319-327 (1970)

5. Drobinski, J. C., Jr., et al. Analysis of Environmental Samples for Carbon^{14} and Tritium. Health Physics, Volume 11, 1965, Pages 385-95.

(c) Strontium-90 in Water

Beta Particle Counting Method

A. Principle

Applicable to H_2O, and to sewage and industrial waste if organic matter is destroyed and interfering ions are eliminated. Added carrier Sr along with radionuclides are separated from other radioactive elements and inactive sample solids by precipitation as $Sr(NO_3)_2$ from fuming HNO_3. Sr is finally precipitated as $SrCO_3$, which is dried, weighed, and set aside ca 2 weeks for ingrowth of ^{90}Y. Precipitate is then dissolved and ^{90}Y is prepared for counting by (*a*) extraction by tributyl phosphate and evaporating on planchet, or (*b*) addition of Y carrier and precipitating as oxalate.

Radioactive Ba and Ra which interfere are removed by addition of Ba carrier, Ca interferes with Sr precipitation, but is removed by HNO_3 precipitation and acetone treatment.

B. Apparatus

(a) *Counting pans.*—Stainless steel, ca 50 mm diameter and 7 mm deep.

(b) *Filtration assembly.*—For mounting precipitates for counting. Consists of (*1*) 2-piece filtering apparatus for 2.4 cm filter such as stainless steel filter holder (Interex Corp., 3 Strathmore Rd, Natick, MA 01760, No. 12-103; ICN Pharmaceuticals, Inc., Life Sciences Group, No. 83012), Teflon filter holder, or equivalent. (2) Nylon (Zytel 101) disk with ring for mounting precipitate.

(c) *Film, Mylar.*—To cover precipitates during counting and storage, ca 0.025 cm thick. Available in rolls 1.5" (3.8 cm) wide as manufacturer's No. 92A, E. I. du Pont de Nemours, Electronics Dept, Barley Mill Plaza, Kirk Mill: Mylar Product Information, Wilmington, DE 19805.

(d) *Glass fiber filter paper.*—No. 934-AH, 2.4 cm diameter, available from Whatman, Inc.

(e) *Centrifuge tubes.*—40 mL, heavy duty with short cone bottom and pour-out lip.

(f) *Beta particle counter.*—Low background, shielded, anticoincidence counter. Determine counter efficiency for ^{90}Y as oxalate and ^{89}Sr as carbonate for specific counter and geometry.

C. Reagents

(a) *Dilute acetic acid.*—6*N*. Add 345 mL CH_3COOH to H_2O and dilute to 1 L.

(b) *Ammonium acetate buffer.*—pH 5.5. Dissolve 154 g NH_4CH_3COO in 700 mL H_2O, add 57 mL CH_3COOH, adjust pH to 5.5 with dropwise addition of either CH_3COOH or 6*N* NH_4OH, as necessary, and dilute to 1 L.

(c) *Dilute ammonium hydroxide.*—6*N*. Dilute 400 mL NH_4OH to 1 L with H_2O.

(d) *Barium carrier solution.*—10 mg Ba/mL. Dissolve 19.0 g $Ba(NO_3)_2$ in H_2O and dilute to 1 L.

(e) *Dilute hydrochloric acid.*—6*N*. Add 500 mL HCl to H_2O and dilute to 1 L.

(f) *Fuming nitric acid.*—21*N*. Specific gravity 1.48, 90% HNO_3.

(g) *Dilute nitric acid.*—(*1*) *14N.*—Add 875 mL HNO_3 to H_2O and dilute to 1 L. (*2*) *6N.*—Add 384 mL HNO_3 to H_2O and dilute to 1 L. (*3*) *0.1N.*—Add 6.25 mL HNO_3 to H_2O and dilute to 1 L.

(h) *Oxalic acid solution.*—Saturated. Approximately 11 g $H_2C_2O_4.2H_2O$ in 100 mL H_2O.

(i) *Mixed rare earth carrier solution.*—Dissolve 12.8 g $Ce(NO_3)_3.6H_2O$, 14 g $ZrOCl_2.8H_2O$, and 25 g $FeCl_3.6H_2O$ in 600 mL H_2O containing 10 mL HCl, and dilute to 1 L. (*Caution:* $Ce(NO_3)_3$ is toxic. Wear resistant rubber or plastic gloves.)

(j) *Sodium carbonate solution.*—2*N*. Dissolve 142 g $Na_2CO_3.H_2O$ in H_2O and dilute to 1 L.

(k) *Sodium chromate solution.*—0.5*M*. Dissolve 117 g $Na_2CrO_4.4H_2O$ in H_2O and dilute to 1 L.

(l) *Sodium hydroxide solution.*—6*N*. Dissolve 240 g NaOH in H_2O and dilute to 1 L.

(m) *Strontium carrier solution.*—10 mg Sr/mL. Dissolve 24.16 g $Sr(NO_3)_2$ in H_2O and dilute to 1 L. Standardize by pipetting (in triplicate) 10 mL solution into 40 mL centrifuge tubes and adding 15 mL 2*N* Na_2CO_3. Stir and heat in boiling H_2O bath 15 min. Filter through weighed, fine porosity, fritted glass, 15 mL crucible. Wash with three 5 mL portions H_2O and three 5 mL portions absolute alcohol or acetone, wipe crucible with absorbent tissue, and dry to constant weight at 110° (ca 20 min). Cool in desiccator and weigh.

$$\text{mg Sr/mL} = \text{mg } SrCO_3 \times 0.5935/10$$

(n) *Tributyl phosphate (TBP), equilibrated.*— Shake TBP with equal volume 14*N* HNO_3. Separate and discard lower acid phase.

(o) *Yttrium carrier solution.*—10 mg Y/mL. Dissolve 12.7 g Y_2O_3 in 30 mL HNO_3 by stirring and heating. Add additional 20 mL HNO_3 and dilute to 1 L with H_2O. 1 mL = 34 mg $Y_2(C_2O_4)_3.9H_2O$. Determine exact equivalence as in step (f) or (g) of D. Determination.

D. Determination

(a) *Precipitation as carbonate.*—To 1 L drinking H_2O (or less, but containing ≥ 25 pCi ^{90}Sr) or filtered raw H_2O sample in beaker, add 2.0 mL HNO_3 and mix. Add 2.0 mL each of Ba and Sr carrier solutions and mix well. (Precipitate of $BaSO_4$ will not cause difficulty.) Heat to bp, and add 20 mL 6*N* NaOH and 20 mL 2*N* Na_2CO_3. Stir, and let simmer ca 1 h at 90–95°. Let cool until precipitate has settled (1–3 h). Decant and discard supernate. Transfer precipitate to 40 mL centrifuge tube, centrifuge, and discard supernate.

(b) *Purification as nitrate.*—Cautiously add 4 mL HNO_3 dropwise to precipitate. Heat to bp, stir, and cool under running H_2O. Add 20 mL fuming HNO_3, cool 5–10 min in ice bath, stir, and centrifuge. Discard supernate. Add 4 mL H_2O to residue, stir, and heat to bp to dissolve $Sr(NO_3)_2$. Centrifuge while hot and decant supernate into clean centrifuge tube. Add 2 mL 6*N* HNO_3 to residue, heat to bp, centrifuge while hot, and combine supernate with aqueous supernate. Discard insoluble residue of $BaSO_4$, SiO_2, etc.

Cool combined supernates, add 20 mL fuming HNO_3, cool 5–10 min in ice bath, stir, centrifuge, and discard supernate. Add 4 mL H_2O to precipitate and dissolve by heating, cool, add 20 mL fuming HNO_3, cool 5–10 min in ice bath, stir, centrifuge, and discard supernate. If >200 mg Ca is present in sample, repeat H_2O solution and fuming HNO_3 precipitation.

(c) *Removal of rare earths.*—After last HNO_3 precipitation, invert tube in beaker ca 10 min to drain off most of excess HNO_3. Add 20 mL acetone to precipitate. Stir thoroughly, cool, centrifuge, and discard supernate. Dissolve precipitate of strontium and barium nitrates in 10 mL H_2O and boil 30 sec to remove any remaining acetone.

Add 0.25 mL mixed rare earth carrier solution and precipitate rare earth hydroxides by making solution basic with 6*N* NH_4OH. Digest in boiling H_2O bath 10 min. Cool in ice bath, centrifuge, decant supernate to clean tube, and discard precipitate. Repeat addition of rare earth carrier solution, precipitation, and decantation. Note time as beginning of ^{90}Y ingrowth period.

(d) *Removal of barium.*—Add 2 drops methyl red indicator and then 6*N* CH_3COOH, dropwise with stirring, until solution is red. Add 5 mL acetate buffer solution, heat to bp, and add 2 mL Na_2CrO_4 solution dropwise with stirring. Digest in boiling H_2O bath 5 min. Cool in ice bath, centrifuge, decant supernate into clean tube, and discard residue.

(e) *Precipitation as strontium carbonate.*—Add 2 mL 6*N* NaOH to supernate; then add 5 mL 2*N* Na_2CO_3 and heat to bp. If pH is <9, add additional NaOH solution. Cool in ice bath ca 5 min, centrifuge, and discard supernate. Add 15 mL H_2O to precipitate, stir, centrifuge, and discard wash H_2O. Repeat washing and weigh $SrCO_3$ as in (*1*) or (*2*):

(*1*) Slurry precipitate with small volume H_2O, and transfer to weighed stainless steel pan. Dry under IR lamp, cool, and weigh. (2) Transfer precipitate to weighed paper or glass filter mounted in 2 piece funnel. Let settle by gravity for uniform deposition; then apply suction. Wash precipitate with three 5 mL portions H_2O, three 5 mL portions alcohol, and three 5 mL portions ether or acetone. Dry 15–30 min in 110–125° oven, cool, and weigh.

Store precipitate ≥ 2 weeks to permit ingrowth of ^{90}Y. Separate and count ^{90}Y by (f) or (g).

(f) *Separation by TBP extraction.*—If $SrCO_3$ is weighed in pan, place pan in small funnel in mouth of 60 mL separator and carefully add 1 mL 6*N* HNO_3 dropwise. Tilt pan to empty, and rinse with two 2 mL portions 6*N* HNO_3.

If $SrCO_3$ is weighed on filter, dislodge bulk of precipitate into small funnel in mouth of 60 mL separator. Cautiously add 1 mL 6*N* HNO_3 dropwise to dissolve remaining precipitate. Rinse filter and funnel with two 2 mL portions of 6*N* HNO_3.

Remove pan or filter and add 10 mL fuming HNO_3 to separator through funnel. Remove funnel and add 1 mL Y carrier solution to separator. Add 5.0 mL TBP, shake thoroughly 3–5 min, let separate, and transfer aqueous layer to second 60 mL separator. Add 5.0 mL TBP to second separator, shake 5 min, let separate, and transfer aqueous layer to third 60 mL separator. Combine TBP extracts and wash with two 5 mL portions 14*N* HNO_3. Record time as beginning of ^{90}Y decay. (Combine acid washings with aqueous phase in third separator if second ingrowth of ^{90}Y is desired.)

Back-extract ^{90}Y from combined organic phase with 10 mL 0.1*N* HNO_3 5 min. Either (*1*) repeat TBP extraction as above, beginning "Add 5.0 mL TBP, . . ." and finally back-extracting ^{90}Y into 10 mL 0.1*N* HNO_3 and continue as in (g), beginning "Gradually . . ."; or (2) transfer aqueous phase to 50 mL beaker and evaporate on hot plate to 5–10 mL. Transfer residual solution to weighed stainless steel counting pan and evaporate. Rinse with two 2 mL portions 0.1*N* HNO_3, add rinsings to counting pan, evaporate to dryness, and weigh. Count in internal proportional or end window counter and calculate ^{90}Sr as in E.

(g) *Separation by yttrium oxalate precipitation.*— Dissolve $SrCO_3$ by cautiously adding 2 mL 6*N* HNO_3 dropwise, and ransfer to 40 mL centrifuge tube, rinsing with 0.1*N* HNO_3. Add 1 mL Y carrier solution, 2 drops methyl red, and NH_4OH dropwise to methyl red end point. Add additional 5 mL NH_4OH and record time as end of ^{90}Y ingrowth and beginning of decay. Centrifuge and decant supernate into beaker. (Save supernate and washings for second ingrowth, if desired.) Wash precipitate with two 20 mL portions hot H_2O. Add 5–10 drops 6*N* HNO_3, stir to dissolve precipitate, add 25 mL H_2O, and heat in H_2O bath at 90°. Gradually add 15–20 drops saturated oxalic acid solution with stirring, and adjust pH to 1.5–2.0 (pH meter or indicator paper) with dropwise addition of NH_4OH. Digest precipitate 5 min; then cool in ice bath with occasional stirring.

Transfer precipitate to weighed glass fiber filter in 2-piece funnel. Let precipitate settle by gravity for uniform deposition and then apply suction. Wash precipitate with 10–15 mL H_2O, three 5 mL portions alcohol, and then three 5 mL portions ether. Air dry precipitate 2 min with suction, weigh, mount on nylon disk and ring with Mylar cover, count, and calculate ^{90}Sr as in E.

E. Calculations

(a) *Strontium-90 calculation.*—

$$^{90}\text{Sr, pCi/L} = \text{net cpm}/(abcdfg \times 2.22)$$

where a = counting efficiency for ^{90}Y; b = chemical yield (fraction) of extracted or precipitated ^{90}Y; c = mg final Y oxalate precipitate/mg Y oxalate in 1 mL carrier; d = chemical yield (fraction) of Sr determined as in D(e) (20 mg Sr equivalent to 33.6 mg $SrCO_3$) or by flame photometry; f = volume, L, original sample; g = ^{90}Y decay factor = $e^{-\lambda t}$; e = base of natural logarithms; $\lambda = 0.693/T_{1/2}$; $T_{1/2}$ = 64.2 h for ^{90}Y; and t = time, h, between separation and counting.

(b) *Counting efficiency.*—Prepare curve from various weights yttrium oxalate precipitate spiked with ^{90}Sr/^{90}Y, precipitated as in D(g).

(c) *Correction for carrier recovery.*—If sample contains more than trace stable Sr, it will act as carrier and will result in >100% yield. In such case determine Sr by flame photometry.

References: JAOAC **56**, 208(1973).
OFFICIAL METHODS of ANALYSIS, AOAC, 15th EDITION, ARLINGTON 1990, Method 973.66

(d) Strontium-89 and Strontium-90 in Milk
Ion Exchange Method

A. Principle

Fresh milk samples are preserved with HCHO and stored to obtain ^{90}Y ingrowth. After storage, Y, Sr, and Ba carriers and citrate solution are added. Citrate forms Y complex which is adsorbed on anion exchange resin. Y is desorbed and separated from radionuclides by tributyl phosphate extraction. Y is re-extracted into dilute HNO_3 and precipitated as oxalate, which is weighed and counted for ^{90}Y activity to calculate ^{90}Sr.

Radio-Sr is desorbed along with Ca and radio-Ba; Ca, radio-Ba, and rare earth radionuclides are separated by repetitive precipitations; Sr is precipitated as $SrCO_3$ and counted. Total radio-Sr minus ^{90}Sr by ^{90}Y measurement yields value for ^{89}Sr.

B. Operating Notes

Radio-Ba and La radionuclides will interfere without purification. Purification from Ca is important for recovery tests but need not be as thorough if Sr recovery is determined by ^{85}Sr tracer or flame photometry. Thoroughly desorb columns before re-use and test periodically to assure complete desorption.

C. Apparatus

See **(A) Strontium-90 in Water B. Apparatus (b)–(f),** plus following:

Ion exchange system.—Consists of 1 L graduated separator, 250 mL separator with fritted glass disk as cation exchange column, and 30 mL separator with fritted glass disk as anion exchange column (Kontes Glass Co., No. K-427530).

D. Reagents

See **Strontium-90 in Water C. Reagents (c), (e)–(g),** plus following:

(a) *Ammonium acetate buffer.*—pH 5.0. Dissolve 153 g NH_4CH_3COO in 900 mL H_2O. Adjust pH to 5.0 with CH_3COOH, using pH meter, and dilute to 1 L.

(b) *Anion exchange resin.*—Dowex 1-X8 (Cl form), analytical grade, 50–100 mesh size, available from Bio-Rad Laboratories.

(c) *Barium carrier solution.*—20 mg Ba/mL. Dissolve 38.1 g $Ba(NO_3)_2$ in H_2O, add 1 mL HNO_3, and dilute to 1 L.

(d) *Cation exchange resin.*—Dowex 50W-X8 (Na form), analytical grade, 50–100 mesh size, available from Bio-Rad Laboratories. Convert commercially available H form into Na form by passing 1.5 L 4*N* NaCl through 170 mL resin placed in column and rinsing with ca 500 mL H_2O until wash H_2O is Cl-free when tested with 1% $AgNO_3$.

(e) *Citrate solution.*—2*M*. Dissolve 384 g anhydrous citric acid (420 g monohydrate) in H_2O, adjust to pH 6.5 with dilute NaOH solution, and dilute to 1 L.

(f) *Oxalic acid solution.*—2*N*. Dissolve 126 g $H_2C_2O_4.H_2O$ in warm H_2O, cool, and dilute to 1 L.

(g) *Silver nitrate solution.*—1%. Dissolve 1 g $AgNO_3$ in H_2O and dilute to 100 mL. Store in brown bottle.

(h) *Sodium chloride solution.*—4*N*. Dissolve 236 g NaCl in H_2O and dilute to 1 L.

(i) *Sodium carbonate solution.*—3*N*. Dissolve 159 g Na_2CO_3 in H_2O and dilute to 1 L.

(j) *Sodium chromate solution.*—1*N*. Dissolve 81 g Na_2CrO_4 in H_2O and dilute to 1 L.

(k) *Strontium carrier solution.*—20 mg Sr/mL. Dissolve 48.3 g $Sr(NO_3)_2$ in H_2O, add 1 mL HNO_3, and dilute to 1 L. Standardize by pipetting 1 mL portions into six separate 40 mL centrifuge tubes containing 15 mL H_2O. Adjust pH (indicator paper or meter) to 8.5–9.0 with 6*N* NH_4OH. Add, with stirring, 3–5 mL 3*N* Na_2CO_3 and digest 5 min in near boiling H_2O bath. Cool to room temperature and process precipitate as in **G(d)** or **(e)**.

(l) *Tributyl phosphate (TBP), pre-equilibrated.*— Add 150 mL H_2O and 30 mL 3*N* Na_2CO_3 to 300 mL TBP in 1 L separator. Shake 2–3 min and let separate. Discard lower aqueous phase. Add 150 mL H_2O to separator, shake 2–3 min, and let separate. Discard lower aqueous phase. Add 150 mL 14*N* HNO_3 and shake 5 min. Let separate and discard lower aqueous phase. Repeat 14*N* HNO_3 treatment twice.

(m) *Yttrium carrier solution.*—10 mg Y/mL. Dissolve 12.7 g Y_2O_3 in 50 mL HNO_3 by heating (avoid boiling). Dilute to 900 mL with H_2O, adjust pH to 2.0 with NH_4OH, and dilute to 1 L with H_2O. Standardize by pipetting 1 mL portions into each of six 40 mL centrifuge tubes containing 15 mL H_2O. Add 5 mL 2*N* oxalic acid and adjust pH to 1.5 with 6*N* NH_4OH, using pH meter. Digest in hot H_2O bath 10 min, and cool to below room temperature. Centrifuge and discard supernate. Process precipitate as in **G(d)** or **(e)**.

Use Y_2O_3 of 99.999% purity (Morton-Thiokol Inc., 152 Andover St, Danvers, MA 01923). Material of lower purity may require purification because of radioactive contaminants.

E. Preparation of Sample

Preserve freshly drawn sample with ca 3 mL HCHO solution for each L milk and refrigerate for known period of time up to 2 weeks to allow ^{90}Y ingrowth. Thoroughly mix preserved, stored sample. If homogeneous, transfer 1 L to separator, C. If nonhomogeneous, before transfer, filter through loose bed of Pyrex glass wool to prevent clogging of resin columns.

F. Removal of Radioelements by Ion Exchange

Combine 1.00 mL each of Y, Sr, and Ba carriers with 10 mL citrate solution (e), in small beaker or vial. Using distilled H_2O, transfer quantitatively to 1 L sample in separator, and mix well.

Add 170 mL Dowex 50W-X8 (d), to 250 mL separator filled with H_2O. Add 15 mL Dowex 1-X8 (h), to 30 mL separator filled with H_2O. Connect all separators together in order sample (top), anion column, cation column (bottom), and place beaker to collect effluent. Open stopcocks of sample, anion, and cation separators, in that order, and note time. Control effluent rate at 10 mL/min with anion column stopcock. Check and adjust effluent flow periodically.

Stop flow when milk level reaches top of each resin bed and note time. Record as mean time the average period of effluent flow. This time is taken as beginning of ^{90}Y decay. Do not permit unnecessary delay during elution. Discard eluate.

Connect separator containing 300 mL warm H_2O, continue elution at 10 mL/min as above, and discard. Separate columns.

G. Yttrium-90 Separation, Purification,

Connect separator containing 100 mL 2*N* HCl to top of anion separator. Open upper stopcock and then lower stopcock, and control effluent flow at 2 mL/min. Collect 15 mL eluate. Close both stopcocks and remove top separator. Stir resin thoroughly with glass stirring rod, and rinse into resin column with small volume 2*N* HCl. Reconnect separator, and continue 2*N* HCl elution, collecting total of 70 mL Y eluate. Retain eluate.

Adjust flow rate to 10 mL/min for remaining 30 mL acid to recharge separator. Discard this eluate. Wash resin with ≥100 mL H_2O until Cl-free by $AgNO_3$ test. Separator is ready for next determination.

Add 5 mL 2*N* oxalic acid to retained eluate and adjust pH to 1.5 with 6*N* NH_4OH, using pH meter. Stir, heat to near bp in H_2O bath, cool in ice bath, centrifuge, decant, and discard supernate. Proceed as in (a) or (b), depending on whether ^{140}Ba-^{140}La is absent or present from gamma analysis of sample.

(a) *If fresh fission products are absent.*—Dissolve precipitate in 1 mL 6*N* HCl, add 15 mL H_2O, and filter through Whatman No. 541 paper into 40 mL centrifuge tube. Wash paper, collecting washings in tube, discard paper, and continue as in (c).

(b) *If fresh fission products are present.*—Dissolve precipitate in 10 mL HNO_3; transfer solution to 60 mL separator, washing centrifuge tube with additional 10 mL HNO_3. Add 10 mL equilibrated TBP, (l), shake 2–3 min, let separate, and drain and discard lower acid phase. Add 15 mL 14*N* HNO_3 to separator, shake 2–3 min, let separate, and drain and discard lower acid phase. Repeat 14*N* HNO_3 treatment to remove light lanthanide elements, particularly ^{140}La. Add 15 mL H_2O to separator and shake 2–3 min. Let separate, and drain aqueous phase containing most of Y into 40 mL centrifuge tube. Repeat wash, using 15 mL 0.1*N* HNO_3, adding it to centrifuge tube.

(c) *Preparation of yttrium oxalate.*—Add 5 mL 2*N* oxalic acid to purified Y solution from (a) or (b), and adjust to pH 1.5 with NH_4OH, using pH meter. Digest solution in hot H_2O bath 10 min with occasional mixing. Cool in ice bath, centrifuge, and discard supernate. Separate and count ^{90}Y oxalate as in (d) or (e), standardize carrier by the same technique used for sample, and calculate ^{90}Sr activity from ^{90}Y count as in I(a).

(d) *Filtration method.*—Place 2.8 cm glass fiber filter on stainless steel planchet and weigh together. Transfer tared filter to filter holder, as in Strontium-90 in Water B(b)(*1*), and assemble.

With H_2O spray, quantitatively transfer yttrium oxalate precipitate to filter funnel, using minimum of suction so that precipitate is distributed uniformly over filter area. Increase suction as necessary after most of precipitate is on filter. Wash precipitate with three 10 mL portions warm H_2O, three 5 mL portions alcohol, and three 5 mL portions ether. Continue suction ca 2–3 min. Carefully remove filter, place on original planchet, and let stand at room temperature 10–15 min. Weigh and calculate yield yttrium oxalate (likely $Y_2(C_2O_4)_3 \cdot 9H_2O$) by dividing this weight by weight obtained on standardization of carrier, D(m).

Remove filter from planchet, place on top of nylon disk, cover with piece of Mylar film, place nylon ring over Mylar film, and press ring onto nylon disk. Cut off excess film. Count ^{90}Y activity, without undue delay, in low background anticoincidence beta counter. Repeat counting after 3 days to confirm purity of ^{90}Y

by its rate of decay. Record dates and time of counting.

(e) *Direct dispersion method.*—Wash precipitate twice with 20 mL portions warm H_2O, cool to below room temperature, centrifuge, and discard supernate. Quantitatively transfer precipitate to tared stainless steel dish. Uniformly disperse precipitate over dish bottom and dry under IR lamp to constant weight. Count in β particle counter.

H. Strontium-89 Separation, Purification,

Connect 1 L separator containing 1 L 4*N* NaCl to cation separator. Open upper stopcock and then lower stopcock, and control effluent flow at 10 mL/min. Collect ca 1 L eluate in 2 L beaker, but leave resin covered with 2–3 mL solution. Retain eluate.

Wash cation separator with 500 mL H_2O from top separator at rate of 10 mL/min. Discard wash H_2O. If resin becomes clogged with milk solids, back-wash separator or transfer resin to beaker, agitate with H_2O, and decant.

Dilute retained eluate to 1.5 L with H_2O, heat to 85–90° on hot plate, and add 100 mL 3*N* Na_2CO_3 with gentle stirring. Remove from heat and cool to room temperature. Decant bulk of clear supernate. Quantitatively transfer precipitate to 250 mL centrifuge bottle with H_2O and centrifuge; discard supernate. Add 50 mL H_2O and disperse precipitate. Centrifuge, discard supernate, and repeat. Dry precipitate 4 h in oven at 110°.

Dissolve precipitate with vigorous stirring by adding ca 4 mL 6*N* HNO_3 in small amounts (magnetic stirrer is helpful). Filter through Whatman No. 541 paper into 40 mL graduated centrifuge tube. Rinse bottle with 4 mL 6*N* HNO_3 and pour washing through paper. Discard paper. Add 20 mL 21*N* HNO_3 to filtrate. Stir and cool in ice bath; centrifuge and discard supernate. ($Sr(NO_3)_2$ precipitation is critical in obtaining good recovery of Sr adequately separated from Ca.) Recoveries from single precipitation are as follows:

HNO_3	Sr Rec., %	Ca Rec., %
14*N*	81 ± 4	2.6 ± 0.9
16*N*	98 ± 1.4	11 ± 2
18*N*	100 ± 1.7	51 ± 3

Dissolve precipitate in 5 mL H_2O and adjust to pH 5.0 with NH_4OH, using pH meter. Add 5 mL NH_4CH_3COO buffer. Heat in H_2O bath, add 1 mL 1*N* Na_2CrO_4, and mix well. Digest in bath 5 min. Centrifuge and decant supernate into small beaker. Evaporate to ca 2 mL, add 2 mL 6*N* HNO_3, and transfer to 40 mL centrifuge tube, using one 3 mL H_2O rinse. Add 20 mL 21*N* HNO_3, stir, cool in ice bath, centrifuge, and discard supernate. Add 3 mL H_2O and 5 mL 6*N* HNO_3 to dissolve precipitate. Add 20 mL 21*N* HNO_3, stir, cool in ice bath, centrifuge, and discard supernate. Record time as beginning of ^{90}Y ingrowth.

Dissolve precipitate in few mL H_2O and adjust pH to 8.5–9.0 with 6*N* NH_4OH. Add 3–5 mL 3*N* Na_2CO_3 to precipitate $SrCO_3$. Centrifuge, and discard supernate. Disperse precipitate in 10 mL H_2O, centrifuge, and discard supernate. Separate and count $SrCO_3$ as in (a) or (b):

(a) *Filtration method.*—Proceed as in **G(d)**, but wash precipitate with three 5–10 mL portions H_2O, transfer to original planchet, and dry 30 min in oven at 110°. Cool in desiccator and weigh. Count as in **G(d)**, record time of counting, and calculate ^{89}Sr as in **I**(b).

(b) *Direct dispersion method.*—Wash precipitate twice with ca 10 mL portions H_2O, dispersing precipitate, centrifuge, and decant and discard supernate. Quantitatively transfer precipitate to tared stainless steel dish. Uniformly disperse precipitate over dish bottom, dry 30 min in 110° oven. Cool in desiccator and weigh. Count in β particle counter. Record time of counting and calculate ^{89}Sr as in **I**(b)

I. Calculations

(a) *For strontium-90 activity.*—

$$^{90}Sr \text{ activity, pCi/L} = (cpm \pm \sigma)/R_S R_Y E_Y D_Y I_Y V$$

where cpm = net beta count rate of ^{90}Y

$$\sigma = \sqrt{\frac{N_s}{t_s} + \frac{N_b}{t_b}}$$

N_S = sample count rate

N_b = background count rate
t_S = sample counting time
t_S = background counting time
R_S = fraction Sr carrier recovered
R_Y = fraction Y carrier recovered
E_Y = counter efficiency for ^{90}Y as Y oxalate, cpm/pCi
D_Y = decay correction factor (= $e^{-\lambda t}$, defined in **973.66E(a)**) for ^{90}Y, where t is time of sepg ^{90}Y from ^{90}Sr to time of counting, **974.37G(d)** or **(e)**
I_Y = ingrowth correction factor (= $1-e^{-\lambda t}$) for degree of equilibrium attained during ^{90}Y ingrowth period, where t is time from start of ingrowth period to time of sepg ^{90}Y from ^{90}Sr
V = sample volume, L

(b) *For strontium-89 activity.-*

^{89}Sr activity,

$$pCi/L=\frac{1}{E_sD_s}\left(\frac{N_s+\sigma}{R_sV}-C_s(a_sE'_s+E_YI_Y)\right)$$

where E_S = counter efficiency for ^{89}Sr as $SrCO_3$, cpm/pCi
D_S = decay correction factor (= $e^{-\lambda t}$) for ^{89}Sr, where t is time from sample collection to time of counting

$$\sigma=\sqrt{\frac{N_s}{t_s}+\frac{N_b}{t_b}}$$

R_S = fraction Sr carrier recovered
N_S = net counts/min of observed radio-Sr
V = sample volume, L
C_S = ^{90}Sr activity, pCi/L
a_S = absorption factor for ^{90}Sr as $SrCO_3$ obtained from self-absorption calibration curve. (Self-absorption curves for ^{89}Sr and ^{90}Sr derived by pptg series of carrier $SrCO_3$ concentrations over expected recovery range in presence of constant amount of ^{89}Sr and ^{90}Y-free ^{90}Sr, respectively. Ordinate is ratio of count rate for each thickness to count rate at 0 sample thickness and abscissa is sample weight for given type of sample mount.)
E'_S = counter efficiency for ^{90}Sr as $SrCO_3$, cpm/pCi
E_Y = counter efficiency for ^{90}Y as Y oxalate, cpm/pCi
I_Y = correction factor (= $1-e^{-\lambda t}$) for degree of equilibrium attained during ^{90}Y ingrowth period, where t is time ^{90}Y was separated from $SrCO_3$ to time of counting, **974.37H(a)** or **(b)**

References: JAOAC **56**, 213(1973); **57**, 37(1974).
OFFICIAL METHODS of ANALYSIS, AOAC, 15th EDITION, ARLINGTON 1990, Method 974.37

(e) Strontium-89 and 90 in food and ashed milk

Application

This method is applicable to the analysis of ash from food samples, including dairy products, and biological materials.

Principle of Method

Strontium carrier is added to an aliquot of dissolved ash and the alkaline earth carbonates are precipitated. The precipitate is dissolved in nitric acid and silica is removed by dehydration. Strontium is then precipitated as the nitrate and purified by washing with nitric acid. Any remaining phosphate is removed by conversion of alkaline earth phosphates to carbonates. Barium and radium are removed by chromate precipitation and final purification is made by hydroxide scavenging. Strontium carbonate is precipitated, dissolved in nitric acid; yttrium carrier added and the solution stored for yttrium-90 ingrowth. The yttrium is then separated from the strontium by extraction into TTA from pH 5 solution, back extracted with dilute nitric acid, precipitated as the oxalate, weighed to determine yield and counted for yttrium-90 beta activity. The strontium is precipitated as the carbonate, weighed and counted for total radio-strontium beta activity.

Reagents

Ammonium Acetate Buffer, pH 5
Ammonium Hydroxide, NH_4OH, Concentrated, (15N), 6N
Barium Carrier, Ba^{++} - 20 mg/ml
Ethyl Alcohol, C_2H_5OH - Absolute (100%)
Hydrochloric Acid, HCl - 6N
Methyl Orange Indicator, 0.1% (w/v)
Nitric Acid, HNO_3 - Concentrated, 1.0N, 0.1N
Oxalic Acid, $H_2C_2O_4$ - 2N
Phenolphthalein Indicator, 1% (w/v)
Scavenging Carrier, Fe^{+++}, Ce^{+++}, Zr^{++++}, 10 mg/ml
Sodium Dichromate, $Na_2Cr_2O_7$ - 0.5N
Sodium Carbonate, Na_2CO_3 - 3N
Strontium Carrier, Sr^{++} - 90 mg/ml
2-Thenoyltrifluoroacetone, TTA - 10% in Benzene
Yttrium Carrier, Y^{+++} - 20 mg/ml

Special Apparatus

Low Background Beta Counter

Procedure

1. Add 1.0 ml of strontium carrier to a 250 ml aliquot of dissolved ash in a 600 ml beaker.

2. Add concentrated NH_4OH to the first appearance of a precipitate, heat to near boiling, and add 3N Na_2CO_3 slowly until the effervescence ceases, then add an additional 150 ml. Allow the precipitate to settle, preferable overnight, decant or suction off the supernatant and discard.

3. Transfer the precipitate to a 250 ml centrifuge bottle using H_2O, centrifuge and discard the supernatant.

4. Dissolve the precipitate in a few ml of concentrated HNO_3 and transfer using a minimum of H_2O to a 250 ml beaker and take to dryness on a hot plate.

5. Bake for 20-30 minutes, cool, and dampen the salts with a few ml of concentrated NHO_3. Add 50 ml of hot H_2O and boil for several minutes (3-5 min.).

6. Filter the hot solution through Watman #541 filter paper into a 250 ml beaker, wash the filter paper with warm water and 1N HNO_3. Take the solution to near dryness on a hot plate. (Add 10 ml of concentrated HNO_3 and again take the solution to near dryness.).

7. Slurry the salts with 30 ml 75% HNO_3 and transfer to a 90 ml centrifuge tube using 75 ml 75% HNO_3. Heat in a water bath, cool in an ice bath, centrifuge, and discard the supernatant.

8. Fill the centrifuge tube to the 75 ml mark with 75% HNO_3, mix well, heat in a water bath, cool in an ice bath, centrifuge and discard the supernatant.

9. Dissolve the precipitate in 15 ml of H_2O, adjust the pH with 6N NH_4OH to a phenophtalein endpoint (pH 9), heat in a water bath, cool, centrifuge, and transfer the supernatant to a 40 ml centrifuge tube.

10. Add 10 ml of H_2O and 10 ml of 3N Na_2CO_3 to the precipitate. Heat with occasional stirring in a water bath for 30 minutes, centrifuge and discard the supernatant. Dissolve the precipitate in 2 ml of concentrated HCl and combine with the supernatant from step 9. Any residue is discarded. After passing this point in the analysis, the procedure must be continued past step 15 in the same day.

11. Adjust the pH of the solution with 6N NH_4OH to a methyl orange endpoint about (pH 4.5) and add 10 ml of pH 5.0 ammonium acetate buffer. Warm the solution in a water bath, add 1 ml of 0.5N $Na_2Cr_2O_7$ and digest for several minutes. Add 1 ml of barium carrier and digest for several minutes. Centrifuge and transfer the supernatant to a 40 ml centrifuge tube.

12. Wash the precipitate with 5 ml of H_2O, centrifuge and combine the supernatant with that from step 11. Discard the precipitate.

13. Neutralize the solution to a pH 7 with 6N NH_4OH and add sufficient Na_2CO_3 to precipitate $SrCO_3$ (~5 - 10 ml). Warm, cool, centrifuge, and discard the supernatant.

14. Dissolve the precipitate in 3 ml of concentrated HCl, and add 20 ml of water and heat in a water bath. Add 3 drops each of scavenging Zr, Ce, and Fe carriers, and make the solution alkaline with 6N NH_4OH. Digest for several minutes, centrifuge, and transfer the supernatant to a 40 ml centrifuge tube. Discard the precipitate. This is the beginning of the ingrowth of yttrium-90.

15. Add 5 ml of 3N Na_2CO_3 to the solution, centrifuge, and discard the supernatant. Record the date.

16. Dissolve precipitate in HNO_3, bring up to 25 ml volume with 1N HNO_3 and H_2O. Stir with 1 ml pipette, blow pipette into solution. Pipette 1 ml into 25 ml volume flask. Blow remaining solution from pipette back to test tube. Fill flask to line with HNO_3. Run diluted solution on flame photometer for pre-extraction strontium yield. Add 1 ml yttrium carrier to remaining 24 ml in test tube and store for at least 12 days ingrowth.

17. Transfer the solution to 60 ml separatory funnel, using a minimum of H_2O; neutralize with 6N NH_4OH to methyl orange endpoint, add 10 ml of ammonium acetate buffer and 10 ml of 10% TTA in benzene. Shake for 5 minutes. Record the time, this is the beginning of the decay of yttrium-90.

18. Transfer the aqueous layer to a clean 60 ml separatory funnel and add 10 ml of 10% TTA in benzene. Shake for 5 minutes. Transfer the aqueous phase to a 90 ml centrifuge tube.

19. The organic layers from steps 17 and 18 are combined in one of the separatory funnels. Wash the empty funnel with 10 ml of ammonium acetate buffer and add it to the funnel containing the organic. Shake for 2 minutes and combine the aqueous phase with that from step 18.

20. Add 5 ml of 0.1N HNO_3 to the separatory funnel and shake for 5 minutes. Transfer the aqueous phase to a 40 ml centrifuge tube.

21. Repeat step 20 twice.

22. Heat the solution from steps 20-21 in a water bath, add 10 ml of 2N $H_2C_2O_4$, adjust pH to 1.0 - 1.5 with concentrated NH_4OH, digest for several minutes, centrifuge and discard the supernatant.

23. Add warm H_2O to the precipitate to the 40 ml mark, mix, centrifuge and discard the supernatant.

24. Place a 2.4 cm glass fiber filter paper on a stainless steel planchet and weigh together. Place the filter paper into a filtration apparatus.

25. Take the precipitate up in 10 ml of warm H_2O and transfer using warm H_2O to the filter paper. Wash the precipitate with three 10 ml portions of ethyl alcohol.

26. Carefully place the filter paper back onto the stainless steel planchet and dry in an oven at 125°C for 20 minutes. Cool in a desiccator and re-weigh the planchet and paper. Calculate the chemical yield of yttrium by dividing the weight of the precipitate by the weight of yttrium oxalate obtained from 1 ml of carrier.

27. Transfer the filter paper plus the precipitate to a plastic holder and cover with mylar. Count for the beta activity of yttrium-90 in a low background beta counter. Record the time of counting, to determine the decay factor of yttrium-90 from the time of separation to the counting time.

28. Heat the combined aqueous phases from steps 18 and 19 in a water bath and add 25 ml of 3N Na_2CO_3. Centrifuge and discard the supernatant.

29. Add 40-50 ml of H_2O to the precipitate, mix well, centrifuge, and discard the supernatant.

30. Place a 2.4 cm glass fiber filter paper on a stainless steel planchet and weigh together. Place the filter paper into a filtration apparatus.

31. Transfer the precipitate to the filter paper using H_2O and wash the precipitate with three 10 ml portions of H_2O.

32. Carefully place the filter paper back onto the stainless steel planchet. Place under a heat lamp for 5-10 minutes, cool in a desiccator and re-weight the planchet and paper.

33. Transfer the filter paper plus the precipitate to a plastic holder and cover with mylar. Count for total radio-strontium beta activity in a low-background beta counter. Record the time of counting, to determine the ingrowth factor of yttrium-90 from the time of separation to the time of counting.

34. Re-dissolve the $SrCO_3$ from the filter paper using 1N HNO_3 and make to volume in a 100 ml volumetric flask using 1N HNO_3. Measure the strontium concentration flame photometrically and compute the yield. If step 16a was done, this step is optional.

Calculations

^{90}Sr pCi/kg =

$$\frac{A\times B\times C}{2.2\times D\times E\times F\times G\times H\times I\times J\times K}$$

A = Net counts per minute yttrium-90.
B = Sample volume in liters.
C = Weight of total ash in grams.
D = Yttrium-90 efficiency factor as determined.
E = Yttrium chemical yield.
F = Yttrium-90 decay factor.
G = Yttrium-90 ingrowth factor.
H = Strontium chemical yield.
I = Weight of ash dissolved in grams.
J = Aliquot taken in liters.
K = Weight of sample in kilograms.

^{89}Sr pCi/kg =

$$\frac{1}{D\times E}\times\frac{A\times B\times C}{2.2\times F\times G\times H\times I}-J(K+L\times M)$$

A = Net counts per minute total radiostrontium.
B = Sample volume in liters.
C = Weight of total ash in grams.
D = Strontium-89 efficiency factor as determined.
E = Strontium-89 decay factor.
F = Strontium chemical yield.
G = Weight of ash dissolved in grams.
H = Aliquot taken in liters.
I = Weight of sample in kilograms.
J = pCi/kg of strontium-90.
K = Strontium-90 efficiency factor as determined.
L = Yttrium-90 efficiency factor.
M = Yttrium-90 ingrowth factor.

References

Goldin, A. S., Velten, R. J., and Friskhorn, G. W., "Determination of Radioactive Strontium," Anal. Chem., 31, 1490 (1959).

Murthy, G. K., Jarnagin, L. R., and Goldin, A. S., "A Method for the Determination of Radionuclides in Milk Ash," J. Dairy Sci., 42 1276 (1959).

(f) Radium-228 in Foods and Water

Introduction

This procedure is for the rapid determination of radium-228 in foods and water. The method can be used to determine radium-228 levels as required in the EPA Drinking Water Standards. (Ref 1)

Radium-228, a Beta Emitter, is found in soils and ores containing naturally occurring thorium nuclides. The detection of radium-228 is difficult because the beta energies emitted are weak having E-Max's of 0.04 and 0.014 MeV. Its detection is complicated also by the presence of other radium isotopes, such as radium-226, 223, and 224, which emit energetic alpha particles, and also emit beta radiation from their short lived daughter products. Radium-228 is determined by measuring the ingrowth of its daughter actinium-228 which has a 6.1 hour half-life. This procedure involves the isolation and measurement of the actinium-228 which has an energetic beta of 2.3 MeV maximum.

The method is dependent on the separation of actinium-228 from contaminating radium nuclides as well as lead, polonium and bismuth daughter products. The interference from actinium-227, a daughter of uranium-235, is insignificant as this nuclide has a weak beta of only 0.047 MeV. making it very difficult to detect. The analytical method presented is a rapid determination of the naturally occurring beta emitter actinium-228 and is based on a combination technique of solvent extraction and ion exchange. The procedure is designed for analysis of natural waters and various foods such as fruits, spices, and tea. It is of radiobiologic interest because actinium-228 is also a measure of radium-228, which if found in foodstuffs and ingested, is hazardous due to the high energetic beta of its immediate daughter product actinium-228 and subsequent alpha emitting daughter products. (Ref 2) The method is sensitive to the level of the EPA Drinking Water Standard, 5 picocuries/liter (pCi/l).

Method

The solvent extraction-ion exchange procedure for the separation of actinium is outlined below. The method is suitable for various foods and natural waters. With modifications, it can be applied to other types of materials.

Initial precipitation of actinium with lanthanum fluoride separates actinium from the bulk of the cations except thorium and uranium. To remove most of the uranium the precipitate is treated with dichromate to keep the uranium soluble in the hexavalent state. The solution is passed through an anion exchange column to complete the removal of lead, bismuth, polonium and any remaining uranium. To remove the thorium, which has been carried quantitatively to this point, the solution is extracted with 2-thenoyltrifluoroacetone (TTA)-benzene solution in the presence of aluminum nitrate. As a final decontamination step, the solution is passed through a cation exchange column from which the actinium is selectively eluted by use of citric acid leaving any radium or thorium on the resin.

Reagents

The two resin columns employed were about 20 centimeters in length. The anion exchange column was made from Dowex 1X8 (100 to 200 mesh). The length of the resin bed was approximately 7 centimeters. The anion exchange resin column was washed in 1.8M HCl and the column prepared by washing with several column volumes of 1.8M HCl. The cation column was made by using Dowex 50X8 (100 to 200 mesh). The column was about 7 centimeters in length and it was made up in 0.2M HNO_3 and prepared by washing with several column volumes of 0.2M HNO_3.

Reagents used were:

(a) 2-thenoyltrifluoroacetone in benzene 0.5M;
(b) sodium hydroxide 19M;
(c) lanthanum nitrate carrier - 12 milligrams lanthanum per milliliter;
(d) hydrochloric acid 1.8M;
(e) nitric acid approximately 0.2M;
(f) ammonium hydroxide 1M;
(g) oxalic acid 7%;
(h) citric acid 5% adjusted to pH 5.0;
(i) hydrofluoric acid 1.0M plus nitric acid 1.0M;
(j) aluminum nitrate 2.0M;

(k) saturated potassium dichromate;
(l) concentrated HF;
(m) HCl and
(n) NH_4OH.

Equipment

(a) Resin Columns 20 cm x 1 cm id.
(b) Filtration Apparatus, Stainless Steel
(c) 1" nylon ring and disk mounts, mylar cover
(d) Low-background beta counter
(e) centrifuge tubes, conical, plastic, 50 ml
(f) glass fiber filter paper

Procedure

1. To a sample in solution (Note 1) add one ml lanthanum carrier. Then, enough concentrated hydrofluoric acid (5 milliliters approximately) is added to precipitate the lanthanum as a fluoride. Stir for 10 minutes. Centrifuge, wash and make final collection in 40 ml centrifuge tube. Make all required washings (after each centrifugation) with 1M HF + 1M HNO_3 in this step.

2. To the collected precipitate, add several drops of saturated $K_2Cr_2O_7$ solution and 1 ml concentrated HF. Dilute to approximately 20 ml, and allow to digest for 5 minutes.

3. Centrifuge, discard aqueous fraction, wash with 20-30 ml HNO_3-HF solution. Repeat washings until precipitate is white discarding supernates. Add 20 ml of NaOH reagent and digest for 5 minutes. Centrifuge, discard aqueous phase. (Wash with NaOH if necessary to remove excess of contaminating precipitate.) Add several drops of 1M NaOH to the $Ac(OH)_3$ precipitate and wash with approximately 20 ml of water. Centrifuge and discard supernate.

4. Dissolve the precipitate in approximately 10 ml of 1.8M HCl. Pass the solution through the previously prepared anion exchange column. Collect the HCl solution which passes through the ion exchange column in centrifuge tube. Wash the column with one column volume ($\approx$ 20 ml) of 1.8M HCl and combine with the solution.

5. To the solution from step 4 add concentrated NH_4OH to reprecipitate the actinium-lanthanum as hydroxides. Centrifuge, discard supernate and wash precipitate with water using several drops of 1M NH_4OH; centrifuge and again discard supernate.

6. To the precipitate add no more than 1 drop concentrated nitric acid; add 5 to 10 ml dilute nitric acid, add 1 ml 2.0M aluminum nitrate. Adjust pH to 1.0 with NaOH, if necessary. Transfer to a 125 ml separatory funnel with dilute nitric acid washings. Add 10 ml of TTA-benzene (0.5M) solution. Shake for 5 minutes. Allow layers to separate and discard benzene layer. In cases of gross thorium contamination, repeat extraction step.

7. Add the entire aqueous phase to a previously prepared cation exchange column, making sure funnel stem has been washed with 0.2M HNO_3. Allow the solution to pass through the column and wash column with 1 column volume of 0.2M nitric acid. Discard solution and washings. Elute with 5% citric acid (pH 5.0) discarding the first column volume and collecting the next 30 ml.

8. To the 30 ml eluent from step 7 add 10 to 12 ml of 7% oxalic acid to reprecipitate the lanthanum-actinium as oxalate. Centrifuge, wash with water (adding several drops of 7% oxalic acid). Centrifuge, transfer the precipitate with washings to glass fiber filter paper, wash, dry, weigh and mount the precipitate on a 1" nylon ring and disk. Count on a low background Beta counter. The actinium-228 decay must be followed for several hours with counting, if necessary, the following day.

NOTE 1

To a sample known to contain large amounts of interfering ions the following clean-up steps must be performed first:

A. Add one ml Barium carrier (20 mg/ml) to sample

B. Precipitate barium with 10 ml of ammonium sulfate (50% w/v)

C. After allowing the barium sulfate to settle well, discard the supernatant and place the precipitate in a test tube

D. Add 2M sodium carbonate to the precipitate, heat in water bath, and occasionally stir to help convert barium sulfate to barium carbonate

E. Dissolve barium carbonate in 1M HCl then put to one side for 36 hours for ingrowth (98.3% equilibrium between ^{228}Ra and ^{228}Ac)

Now proceed to Step 1.

References

1. EPA (1976) National Interim Primary Drinking Water Regulations, EPA-570/9-76-003, US EPA, Washington, DC

2. FDA (1979) "Quality Standards for foods with no identity standards- bottled water", Fed. Reg. 44 (45), 12169

3. Baratta, E.J. and M.H. Feldman, "Determination of Actinium in Uranium Mill Effluents", USAEC Topical Report WIN-123 (1961)

4. NBS Technical Note-1137 "Evaluation of Methods for Assay of Radium-228 in Water" National Bureau of Standards U.S. Dept. of Commerce (1981)

5. Baratta, E.J. and Ella M. Lumsden "Determination of Radium-228 in Foods and Water" JOAC 65 No.6 (1982)

(3) Alpha-emitting Radionuclides

(a) Uranium

A. Occurrence

Uranium, the heaviest naturally occurring element, is a mixture of three radioactive isotopes: uranium 238(99.275%), uranium 235(0.72%), and uranium 234(0.005%). Most drinking water sources, especially ground waters, contain soluble carbonates and bicarbonates that complex and keep uranium in solution.

B. Method

This is a radiochemical procedure which determines total uranium alpha activity without making an isotopic uranium analysis.

Radiochemical Method

A. General Discussion

a. Principle: The sample is acidified with hydrochloric or nitric acid and boiled to eliminate carbonate and bicarbonate ions. Uranium is co-precipitated with ferric hydroxide and subsequently separated. The ferric hydroxide is dissolved, passed through an anion-exchange column, and washed with acid, and the uranium is eluted with dilute hydrochloric acid. The acid eluate is evaporated to near dryness, the residual salt is converted to nitrate, and the alpha activity is counted.

b. Interference: The only alpha-emitting radionuclide that may be carried through this procedure is protactinium 231. However, this isotope, which is a decay product of uranium 235, causes very little interference. Check reagents for uranium contamination by analyzing a complete reagent blank.

c. Sampling: Preserve sample by adjusting its pH to <2 with HCl or HNO_3, at time of collection.

B. Apparatus

a. Counting instrument, gas-flow proportional or alpha scintillation counting system.

b. Ion-exchange column, approximately 13 mm ID x 150 mm long with 100-mL reservoir.

c. Membrane filter apparatus, 47-mm diam.

C. Reagents

a. Ammonium hydroxide, NH_4OH, $5N$, 1%
b. Anion-exchange resin.[*]
c. Ferric chloride carrier: Dissolve 9.6 g $FeCl_3 \cdot 6H_2O$ in 100 mL $0.5N$ HCl; 1 mL = 20 mg Fe^{3+}.
d. Hydriodic acid (hydrogen iodide), HI, 47%
e. Hydrochloric acid, HCl, conc, $8N$, $6N$, $0.1N$.
f. Iodic acid, 1mg/mL: Dissolve 100 mg HIO_3 in 100 mL $4N$ HNO_3.
g. Nitric acid, HNO_3, conc, $4N$
h. Sodium hydrogen sulfite, 1%: Dissolve 1 g $NaHSO_3$ in 100 mL $6N$ HCl.
i. Uranium standard solution: Dissolve 177.3 mg natural undepleted uranyl acetate[†], $UO_2(C_2H_3O_2)_2 \cdot 2H_2O$, in 1000 mL $0.2N$ HNO_3; 1 mL = 100 μg U = 150 dpm U = 67.6 pCi U. NOTE: Commonly available uranyl salts may be formed from depleted uranium; verify isotopic composition before use.

[*] Dowex 1X4, 100-200 mesh, chloride form, or equivalent.

[†] Standard radioactive solutions with uranium isotopes in equilibrium are available from the U.S. Environmental Protection Agency, Environmental Monitoring Systems Laboratory, NRA/STD, P.O. Box 93478, Las Vegas, Nev. 89193.

D. Calibration

Determine counting efficiency, *E*, for a known amount of uranium standard solution (about 750 dpm) evaporated from 6 to 8 mL of 1 mg/mL HIO_3 solution in a 50-mm-diam stainless steel planchet. After flaming planchet, count for at least 50 min. Run a reagent blank with the standard portions and count.

$$E = \frac{C - B}{D}$$

where:

C= gross alpha count rate of standard, cpm,
B= alpha background count rate, cpm, and
D= disintegration rate of uranium standard, dpm.

Determine uranium recovery factor by adding a measured amount of uranium standard to the same volume of sample and taking it through the entire procedure. Alpha count the separated, evaporated, and flamed uranium planchet. Determine the recovery factor on at least 10% of all drinking water samples. For non-drinking water samples, it may be necessary to determine the recovery factor in every sample.

$$\text{Recovery factor}, R = \frac{C' \; B'}{DE}$$

where:

C' = gross count rate of sample with added uranium, cpm,
B' = count of reagent blank, cpm,
D = disintegration rate of uranium standard, dpm, and
E = counting efficiency.

E. Procedure

1. If the sample has not been acidified, add 5 mL conc HCl or HNO_3 to 1 L sample in a 1500-mL beaker. Add 1 mL $FeCl_3$ carrier. In each batch of samples include a distilled-water blank. Cover with watch glass and heat to boiling for 20 min. If pH is greater than 1, add conc HCl or HNO_3 dropwise to bring pH to 1. While sample is boiling, gently add 5*N* NH_4OH until turbidity persists while boiling continues; then add 10 mL more. Continue boiling for 10 min more, then set aside for 30 min to cool and settle. After sufficient settling, decant and filter supernate through a 47-mm, 0.45-μm membrane filter using a large filtering apparatus. Slurry the remaining precipitate, transfer to the filtering apparatus, and filter with suction. Complete transfer using 1% solution of NH_4OH delivered from a polyethylene squeeze bottle. Place filtering apparatus over a clean 250-mL filtering flask, add 25 mL 8*N* HCl.

2. Prepare an ion-exchange column by slurrying the anion-exchange resin with 8*N* HCl and pouring it into a 13-mm-ID column to give a resin bed height of about 80 mm. Transfer solution to the 100-mL reservoir of the ion-exchange column. Rinse side-arm filtering flask twice with 25-mL portions of 8*N* HCl. Combine in the ion-exchange reservoir. Pass sample solution through the anion-exchange column at a flow rate of not more than 5 mL/min. After sample has passed through column, elute the iron (and plutonium if present) with six column volumes of freshly prepared 8*N* HCl containing 1 mL 47% HI /9 mL 8*N* HCl. Wash column with two additional column volumes of 8*N* HCl. Discard all washes. Elute uranium into a 100-mL beaker with six column volumes of 0.1*N* HCl. Evaporate acid elute to near dryness and convert residue to the nitrate form by three successive treatments with 5-mL portions of conc HNO_3, evaporating to near dryness each time. *Do not bake.* Dissolve residue (of which there may be very little visible) in 2 mL 4*N* HNO_3. Using a transfer pipet, transfer to a marked planchet. Complete transfer by rinsing beaker three times with 2-mL portions of 4*N* HNO_3. Evaporate planchet contents to dryness under a heat lamp, flame to remove traces of HIO_3, cool, and count for alpha activity.

3. To regenerate anion-exchange resin column, pass three column volumes of 1% $NaHSO_3$ in 6*N* HCl through the column, follow with six column volumes of 6*N* HCl, and then three column volumes of distilled water. Do not let resin become dry. When ready for the next set of samples, equilibrate by passing six column volumes of 8*N* HCl through the column.

F. Calculations

$$\text{Uranium } \alpha \text{ activity, pCi/L} = \frac{C'' - B'}{2.22 \times ERV}$$

where:

C'' = gross count rate of sample, cpm,
V = volume of sample, L, and other factors are as defined above.

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STANDARD METHODS FOR THE EXAMINATION OF WATER AND WASTE WATER, *APHA 18th Edition* (1992)

(b) Radium-226

Precipitation Method

A. General Discussion

a. Application: This method is suitable for determination of the alpha-emitting isotopes of radium.

b. Principle: Because of the difference in half-lives of the nuclides in the series including the alpha-emitting Ra isotopes, these isotopes can be identified by the rate of ingrowth and decay of their daughters in a barium sulfate precipitate.[1-3] The ingrowth of alpha activity from radium 226 increases at a rate governed primarily by the 3.8-d half-life radon 222. The ingrowth of alpha activity in radium 223 is complete by the time a radium-barium precipitate can be prepared for counting. The ingrowth of the first two alpha-emitting daughters of radium 224 is complete within a few minutes and the third alpha daughter activity increases at a rate governed by the 10.6-h half-life of lead 212. The activity of the radium 224 itself, with a 3.6-d half-life, also is decreasing, leading to a rather complicated ingrowth and decay curve.

Lead and barium carriers are added to the sample containing alkaline citrate, then sulfuric acid (H_2SO_4) is added to precipitate radium, barium, and lead as sulfates. The precipitate is purified by washing with nitric acid (HNO_3), dissolving in alkaline EDTA, and re-precipitating as radium-barium sulfate after pH adjustment to 4.5. This slightly acidic EDTA keeps other naturally occurring alpha emitters and the lead carrier in solution.

B. Apparatus

a. Counting instruments: One of the following is required:
1) *Internal proportional counter*, gas-flow, with scaler and register.
2) *Alpha scintillation counter*, silver-activated zinc sulfide phosphor deposited on thin polyester plastic, with photomultiplier tube, scaler, timer, and register; or
3) *Proportional counter*, thin end-window, gas-flow, with scaler and register.
b. Membrane filter holder, or stainless steel or TFE filter funnels, with vacuum source (Fisher Filtraton or equiv.).
c. Membrane filters (Millipore Type HAWP or equiv.), or *glass fiber filters* (No. 934-AH, diam. 2.4 cm, H. Reeve Angel or equiv.).

C. Reagents

a. Citric acid, 1M: Dissolve 210 g $H_3C_6H_5O_7.H_2O$ in distilled water and dilute to 1 L.
b. Ammonium hydroxide, conc and 5*N*: Verify strength of old 5*N* NH_4OH solution before use.
c. Lead nitrate carrier: Dissolve 160 g $Pb(NO_3)_2$ in distilled water and dilute to 1 L; 1 mL = 100 mg Pb.
d. Stock barium chloride solution: Dissolve 17.79 g $BaCl_2.2H_2O$ in distilled water and dilute to 1 L in a volumetric flask; 1 mL = 10 mg Ba.
e. Barium chloride carrier: To a 100-mL volumetric flask, add 20.00 mL stock $BaCl_2$ solution using a transfer pipet, dilute to 100 mL with distilled water, and mix; 1 mL = 2.00 mg Ba.
f. Methyl orange indicator solution.
g. Phenolphthalein indicator solution
h. Bromcresol green indicator solution: Dissolve 0.1 g bromcresol green sodium salt in 100 mL distilled water.
i. Sulfuric acid, H_2SO_4, 18*N*.
j. Nitric acid, HNO_3, conc.
k. EDTA reagent, 0.25*M*: Add 93 g disodium ethylenediaminetetraacetate dihydrate to distilled water, dilute to 1 L, and mix.
l. Acetic acid, conc.
m. Ethyl alcohol, 95%.
n. Acetone.
o. Clear acrylic solution: § Dissolve 50 mg clear acrylic in 100 mL acetone.

§Lucite or equivalent.

p. Stock radium 226 solution: Take every precaution to avoid unnecessary contamination of working area, equipment, and glassware, preferably by preparing ^{226}Ra standards in a separate area or room reserved for this purpose. Obtain a National Institute of Standards and Technology (NIST, formely National Bureau of Standards, NBS or other equivalent source ie; National Physics Laboratory U.K.; Institute for Reference Materials and Measurements, E.U.) gamma ray standard containing 0.1 μg ^{226}Ra as of date of standardization. Using a heavy glass rod, cautiously break neck of ampule, which is submerged in 300 mL acid $BaCl_2$ solution in a 600-mL beaker. Chip ampule unit until it is thoroughly broken or until hole is large enough to give complete mixing. Transfer solution to a 1-L volumetric flask, rinse beaker with acid $BaCl_2$ solution, dilute to mark with same solution, and mix; 1 mL = approximately 100 pg ^{226}Ra. Determine the time in years, t, since the NIST or other standardization of the original ^{226}Ra solution. Calculate pCi ^{226}Ra/mL as:
pCi ^{226}Ra = $[1-(4.3\text{x}10^{-4})(t)][100][0.990]$
q. Intermediate radium 226 solution: Dilute 100 mL stock radium solution to 1000 mL with acid $BaCl_2$ solution: 1 mL = approximately 10 pCi ^{226}Ra.
r. Standard radium 226 solution: Add 30.0 mL intermediate radium solution to a 100-mL volumetric flask and dilute to mark with acid $BaCl_2$ solution: 1 mL = approximately 3 pCi ^{226}Ra and contains about 2 mg of Ba. See ¶*p et seq.* above for corrections.

D. Procedure for Radium in Drinking Water and for Dissolved Radium

1. To 1 L sample in a 1500-mL beaker, add 5 mL 1*M* citric acid, 2.5 mL conc NH_4OH, 2 mL $Pb(NO_3)_2$ carrier, and 3.00 mL $BaCl_2$ carrier. In each batch of samples include a distilled water blank.
2. Heat to boiling and add 10 drops methyl orange indicator.
3. While stirring, slowly add 18*N* H_2SO_4 to obtain a permanent pink color; then add 0.25 mL acid in excess.
4. Boil gently 5 to 10 min.
5. Set beaker aside and let stand until precipitate has settled (3 to 5 h or more). ‖

‖If original concentrations of isotopes of radium other than ^{226}Ra are of interest, note date and time of this original precipitation as the separation of the isotopes from their parents: use a minimal settling time and complete procedure through ¶*i* without delay. Assuming the presence of and separation of parents, decay of ^{223}Ra and ^{224}Ra begins at the time of the first precipitation(¶*i*). The time of the first precipitation is not needed if the objective is to check the final precipitate for its ^{226}Ra content only.

6. Decant and discard clear supernate. Transfer precipitate to a 40-mL or larger centrifuge tube, centrifuge, decant, and discard supernate.
7. Rinse wall of centrifuge tube with a 10-mL portion of conc HNO_3, stir precipitate with a glass rod, centrifuge, and discard supernate. Repeat rinsing and washing two more times.
8. To precipitate, add 10 mL distilled water and 1 to 2 drops phenolphthalein indicator solution. Stir and loosen precipitate from bottom of tube (using a glass rod if necessary) and add 5*N* NH_4OH, dropwise, until solution is definitely alkaline (red). Add 10 mL EDTA reagent and 3 mL 5*N* NH_4OH. Stir occasionally for 2 min. Most of the precipitate should dissolve, but a slight turbidity may remain.
9. Warm in a steam bath to clear solution (about 10 min.), but do not heat for an unnecessarily long period.[#] Add conc acetic acid dropwise until red color disappears; add 2 or 3 drops bromcresol green indicator solution and continue to add conc acetic acid dropwise, while stirring with a glass rod, until indicator turns green (aqua).[##] $BaSO_4$ will precipitate. Note date and time of precipitation as zero time for ingrowth of alpha activity. Digest in a steam bath for 5 to 10 min, cool, and centrifuge. Discard supernate. The final pH should be about 4.5, which is sufficiently low to destroy the Ba-EDTA complex, but not Pb-EDTA. A pH much below 4.5 will precipitate $PbSO_4$.

[#] If solution does not clear in 10 min. cool, add another mL 5*N* NH_4OH, let stand 2 min. and heat for another 10-min period.
[##] The end point is most easily determined by comparison with a solution of similar composition that has been adjusted to pH 4.5 using a pH meter.

10. Wash Ba-Ra sulfate precipitate with distilled water and mount in a manner suitable for counting as given in ¶'s *11, 12 or 13* following.
11. Transfer Ba-Ra sulfate precipitate to a tared stainless steel planchet with a minimum of 95% ethyl alcohol and evaporate under an infrared lamp. Add 2 mL acetone and 2 drops clear acrylic solution, disperse recipitate evenly, and evaporate under an infrared lamp. Dry in oven at 110°C, weigh, and

determine alpha activity, preferably with an internal proportional counter. Calculate net counts per minute and weight of precipitate.

12. Weigh a membrane filter, a counting dish, and a weight (glass ring) as a unit. Transfer precipitate to tared membrane filter in dish, add glass ring, and dry at 110°C. Weigh and count in one of the counters mentioned under ¶ B*a* above. Calculate net counts per minute and weight of precipitate.
13. Add 20 mL distilled water to the Ba-Ra sulfate precipitate, let settle in a steam bath, cool, and filter through a special funnel with a tared glass fiber filter. Dry precipitate at 110°C to constant weight, cool, and weigh. Mount precipitate on a nylon disk and ring with an alpha phosphor on polyester plastic film[4], and count in an alpha scintillation counter. Calculate net counts per minute and weight of precipitate.
14. If the isotopic composition of the precipitate is to be estimated, perform additional counting as mentioned in the calculation below.
15. *Determination of combined efficiency and self-absorption factor:* Prepare standards from 1 L distilled water and the standard radium 226 solution (¶ C*p* preceding). Include at least one blank. The barium content will impose an upper limit of 3.0 mL on the volume of the standard radium 226 solution that can be used. If x is volume of standard radium 226 added, then add (3.00-x)mL $BaCl_2$ carrier(¶3*e* above). Analyze standards as samples, beginning with ¶D*1*, but omit 3.00-mL $BaCl_2$ carrier.

From the observed net count rate, calculate the combined factor, *bc*, from the formula:

$$bc = \frac{net\ cpm}{ad \times 2.22 \times pCi\ radium\ 226}\dagger\dagger$$

where:

ad = ingrowth factor (see below) multiplied by chemical yield.

††See calculation that follows

If all chemical yields on samples and standards are not essentially equal, the factor *bc* will not be a constant. In this event, construct a curve relating the factor *bc* to varying weights of recovered $BaSO_4$.

5. Calculation

$$Radium,\ pCi/L = \frac{net\ cpm}{a\ b\ c\ d\ e \times 2.22}$$

where:

a = ingrowth factor (as shown in the following tabulation):

Ingrowth *h*	Alpha Activity from ^{226}Ra
0	1.000
1	1.016
2	1.036
3	1.058
4	1.080
5	1.102
6	1.124
24	1.489
48	1.905
72	2.253

b = efficiency factor for alpha counting
c = self-absorption factor
d = chemical yield
e = sample volume

The calculations are based on the assumption that the radium is radium 226. If the observed concentration approaches 3 pCi/L, it may be desirable to follow the rate of ingrowth and estimate the isotopic content[2,3] or, preferably, to determine radium 226 by radon 222.

The optimum ingrowth periods can be selected only if the ratios and identities of the radium isotopes are known. The number of observed count rates at different ages must be equal to or greater than the number of radium isotopes present in a mixture. In the general case, suitable ages for counting are 3 to 18 h for the first count; for isotopic analysis, additional counting at 7, 14, or 28 d is suggested, depending on the number of isotopes in mixture. The amounts of the various radium isotopes can be determined by solving a set of simultaneous equations.[3] This approach is most satisfactory when radium 226 is the predominant isotope; in other situations, the approach suffers from statistical counting errors.

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5. STANDARD METHODS FOR THE EXAMINATION OF WATER AND WASTE WATER, *APHA 18TH Edition(1992)*

(c) Isotopic Analysis of Pu in Food Ash

The method was set up for soil and has been changed slightly to use on food ash. The method includes a procedure for (A) 10 gram ash samples and (B) a rapid procedure for 1 gram ash sample.

I. Procedure A

Summary

The principle of the analytical procedure follows: A known quantity of plutonium-236 tracer is added to the ashed sample which is decomposed by sequential nitric acid-hydrofluoric acid and nitric acid-hydrofluoric acid-hydrochloric acid digestions. Boric acid is added to complex the fluoride ion and to aid in the extraction of plutonium from any remaining insoluble residue. Sequential iron hydroxide precipitations are performed with sodium hydroxide and ammonium hydroxide to respectively remove amphoteric elements and calcium and to eliminate soluble fluorides. The hydroxide precipitate is dissolved in nitric acid saturated with boric acid and plutonium is isolated and purified by ion exchange separation. The plutonium is then electroplated on a stainless steel disk. The plutonium isotope and concentrations are determined by alpha spectrometry.

Tracer

Plutonium-236 solution - stock standard solution of 2.5×10^4 dpm of plutonium-236 in 2 M nitric acid in minimal solution (available from Amersham Corp). Solution made for tracer should contain approximately 10 dpm/ml.

Acids

All solutions are made with distilled water. All acids are reagent grade and meet American Chemical Society (ACS) specifications.

(a) Boric Acid - crystalline
(b) Hydrochloric Acid - concentrated (12 M), 4 M and 2 M
(c) Hydrofluoric Acid - concentrated (48% solution)
(d) Nitric Acid - concentrated (16 M), 8 M saturated with boric acid, 8 M

Reagents

All solutions are made with distilled water. All reagents are reagent grade and meet ACS specifications unless otherwise defined.

(a) Ammonium Hydroxide - concentrated (15 M)
(b) Ammonium Iodide-Hydrochloric Acid Solution - 1 part of 1 M ammonium iodide to 20 parts 12 M hydrochloric acid (freshly made)
(c) Anion Exchange Resin - Dowex 1X4 (100-200 mesh, nitrate or chloride form)
(d) Iron Carrier - 10 mg Fe (III)/ml in 2 M hydrochloric acid
(e) Sodium Bisulfite - anhydrous
(f) Sodium Nitrite - crystalline
(g) Sodium Hydroxide - 50% solution

Instrumentation

1. A windowless 2 pi gas flow proportional counter.

2. An alpha spectrometer system capable of 40 to 50 KeV resolution of actual samples electrode deposited on flat, minor finished stainless steel planchets with a counting efficiency of >17% and a background of <0.010 cpm over the designated energy region. Resolution is defined as the full width half maximum (FWHM) in KeV, the distance those points on either side of the alpha peak where the counter is equal to one-half the maximum count (Heath, 1964)

Anion Exchange Resin Columns -- Dowex 1X4 (100-200 mesh nitrate form)

Remove the fines from an appropriate amount of resin by repeated suspension in distilled water and decantation. Decant the water and add a volume of concentrated (16 M) nitric acid approximately equal to 50% of the volume of the resin slurry. Using 8 M nitric acid from a wash bottle, transfer sufficient resin to a 1.3 cm ID ion exchange column to give a 10-cm bed of settled resin. Convert the resin to the nitrate form by passing 100 ml of 8 M nitric acid through the column at maximum flow rate.

Sample Decomposition

1. Weigh a representative 10.0 ± 0.1 gram of the food ash sample* and transfer to a 250 ml PTFE (Teflon) beaker.

   * Sample weight is approximately 200 g before drying and ashing.

2. Add 16 M nitric acid, a few drops at a time, as fast as the frothing and vigor of the reaction will permit until the entire sample is covered.

3. Add an appropriate quantity of plutonium-236 tracer.

   Note: If the activity is expected to be less than 1 dpm/g, or is unknown, add 10 dpm of the tracer. For higher levels add as much plutonium-236 tracer as the estimated activity of plutonium-239 or plutonium-238 in the sample.

4. Add an additional 60 ml of 16 M nitric acid and 30 ml of 48% hydrofluoric acid and digest on a hotplate with frequent stirring (Teflon stirring rod) for about 1 hour.

   Caution: Hydrofluoric acid is an extremely hazardous liquid. Use gloves to avoid contact with skin and work in a fume hood to avoid breathing vapors.

5. Remove from the hotplate and cool somewhat before adding 30 ml each of 16 M nitric acid and 48% hydrofluoric acid. Digest on the hotplate with intermittent stirring for an additional hour.

6. Remove from the hotplate and cool completely. Carefully add 20 ml of 12 M HCl and stir. Heat on a hotplate for 45 minutes with occasional stirring after initial reaction has subsided.

7. Add 5 g of powdered boric acid and digest for an additional 15 minutes with occasional stirring.

8. Add approximately 200 mg of sodium bisulfite crystals and continue heating until the solution has evaporated to a liquid volume of approximately 10 ml.

9. Add 50 ml of distilled water and digest on a hotplate with stirring for 10 minutes to dissolve the soluble salts. (Some insoluble salts may remain.)

Sodium Hydroxide Precipitation

10. Cool and transfer approximately equal parts of the total sample (including insolubles if present) into two 250 ml centrifuge bottles with a minimum of distilled water from a wash bottle.

11. Add 1 ml of iron carrier solution (10 mg Fe^{3+}/ml) to each centrifuge bottle and stir.

12. Add 50% sodium hydroxide with stirring to each bottle to a pH of approximately 9. Add 5-10 ml excess of the sodium hydroxide and stir for 1 minute.

13. Centrifuge for approximately 5 minutes, decant and discard the supernate(s).

14. Dissolve each precipitate with about 30 ml (60 ml total) of 8 M nitric acid saturated with boric acid. Digest on a hot water bath for 10 minutes.

15. Cool and centrifuge for 5 minutes. Decant the supernates into the original 250 ml PTFE beaker and save.

16. Wash each residue with approximately 10-20 ml (20-40 ml total) of 8 M nitric acid saturated with boric acid. Centrifuge for 5 minutes and combine the supernates with those in Step 15. (Discard remaining residue).

17. Heat the supernates on a hotplate and evaporate to near dryness.

Ammonium Hydroxide Precipitation

18. Add approximately 30 ml of water and heat to dissolve the salts. Cool, and transfer equal portions into centrifuge bottles.

19. Add 15 M ammonium hydroxide dropwise with stirring to a pH of approximately 9. Centrifuge and discard the supernate.

Ion Exchange Separations

20. Dissolve the precipitate(s) with a volume of 16 M nitric acid approximately equal to the volume of the precipitate(s) and transfer using 8 M nitric acid into a 250 ml beaker. Add 8 M nitric acid to a total volume of approximately 75 ml.

    Note: If the volume of the hydroxide precipitate is considerably greater than should be expected from the 10 mg of Fe added, the final volume should be brought up to approximately 100 ml with 8 M nitric acid or, alternatively, the dissolved hydroxides should be evaporated to salts before addition of the 8 M nitric acid solution. The final molarity of the HNO_3 is not extremely critical, but should be in the range of 7-9.

21. Add approximately 200 mg of sodium nitrite ($NaNO_2$) crystals and stir with a stirring rod. Bring to a quick gentle boil on a hot plate, and cool. Avoid prolonged heating.

22. Pass the sample (at maximum flow rate) through an anion-exchange resin column (nitrate form) prepared as described previously.

23. When the solution just drains to the top of the resin bed, add 8 M nitric acid to the column reservoir and wash the resin column at maximum flow rate. Discard the effluents from the sample and washes.

24. Repeat step 23 until the resin column has been washed with a total of six column volumes of 8 M nitric acid.

25. Wash the resin column with six column volumes of 12 M hydrochloric acid using the same technique. Discard the hydrochloric acid washes.

26. Elute the plutonium with four column volumes of freshly prepared ammonium iodide-hydrochloric acid solution (1 part 1 M NH_4I to 20 parts of 12 M HCl) and collect in a 150 ml beaker.

27. Evaporate the solution to approximately 5 ml on a hot plate. Add 2 ml of 16 M nitric acid after reaction stops. Add 1 ml $HClO_3$. When fumes start add 2 ml conc H_2SO_4 and evaporate down to a 1 ml of H_2SO_4.

Preparation and Plating of Sample

28. To the beaker containing the sample, add 0.5 ml H_2O. Swirl beaker and transfer the activity to the cell using a medicine dropper.

29. Add 7 drops of 0.7 M H_2SO_4 and approximately 0.5 ml H_2O to the beaker. Swirl and transfer with a dropper to the cell.

30. Wash the beaker with approximately 0.5 ml H_2O once or twice and transfer to the cell with a dropper.

31. Add indicator methyl orange or methyl red to the cell.

32. Add to the cell one dropperful of NH_4OH. Using automatic stirrer, titrate to the color change with NH_4OH. Back titrate with 0.7 M H_2SO_4 to the color change. Add 2 drops in excess. The volume should not be much above the stirrer, approximately 7 ml.

33. Plate for 2 hours at approximately 450 milliamps, between 5 and 6 volts. Then add one dropperful NH_4OH and plate for 1 minute. Pour supernate into a beaker and save until plating has been known

to be successful.

34. Disassemble the cell. Rinse plate in a beaker of H_2O containing some NH_4OH and then ignite in a flame.

Calculations

$$\text{pCi/g} = \frac{A}{2.2 \text{x} B \text{x} C \text{x} D}$$

B = Instrument counting efficiency (Alpha spectrometer/Silicone diode detector)

$$B = \frac{R_S \text{ x } E_{2\pi}}{R_{2\pi}{}^{*}} \quad ({}^{*}2\pi = 2 \text{ pi})$$

C = Recovery factor

$$C = \frac{R_6}{T \text{ x } B}$$

D = Weight in grams of sample

R_S = the net counting rate of the known electroplated source over the entire energy region on the alpha spectrometer (cpm)

$E_{2\pi}$ = the counting efficiency of the 2π counter

$R_{2\pi}$ = the net counting rate of the same source (R_S) on the 2 pi counter in cpm

R_6 = net count rate in ^{236}Pu energy region

T = time

B = efficiency

II. Procedure B

The principle of the analytical procedure for 1 g sample follows (analysis of effluent and environmental samples from uranium mills and of biological samples - U. S. Public Health Service, D. R. Rushing, J. Garcia and D. A. Clark).

A known amount of Pu-236 tracer is added to the sample which is decomposed by a flux which in turn is dissolved in 3 M H_2SO_4. The activity is then precipitated, as LaF_3 which in turn is dissolved in 2 M aluminum nitrate and 2 M HNO_3. Hydroxylamine hydrochloride is added and then sodium nitrite to have the Pu all in the same state. The solution is purified by TTA. Four samples tried - 2 tea and 2 pepper. Average mean recovery was 77.0 ± 5.0

Tracer

Plutonium-236 solution - 2.5 x 10^4 dpm of plutonium-236 in a 2 M nitric acid in minimal solution (available from Amersham Corp). Solution made up for tracer should contain approximately 10 dpm/ml.

Acids

All solutions are made with distilled water. All acids are reagent grade and meet the American Chemical Society (ACS) specifications.

(a) Sulfuric Acid, concentrated
(b) Hydrofluoric Acid, concentrated and 1 M
(c) Nitric Acid 10 M, 2 M and 1 M
(d) Perchloric Acid, concentrated

Reagents

All solutions are made in distilled water unless noted. All reagents are reagent grade and meet ACS specifications unless defined otherwise.

(a) TTA in benzene, 0.45 M.
(b) Aluminum Nitrate, 2 M
(c) Hydrogen Peroxide, 30%
(d) Hydroxylamine Hydrochloride, 1 M
(e) Sodium Nitrate, crystalline
(f) Sodium Carbonate, anhydrous
(g) Sodium Tetraborate, powdered
(h) Lanthanium Nitrate, 20 mg/ml

Instrumentation

1. A windowless 2 pi gas flow proportional counter.

2. An alpha spectrometer system capable of 40 to 50 KeV resolution of actual samples electrode deposited on flat, minor finished stainless steel planchets with a counting efficiency of >17% and a background of <0.010 cpm over the designated energy region. Resolution is defined as the full width half maximum (FWHM) in KeV, the distance those points on either side of the alpha peak where the counter is equal to one-half the maximum count (Heath, 1964).

Procedure for a 1 g Ash Sample for Plutonium in Food

1. Use Nicholson's original flux - sodium carbonate and sodium tetrabonate in a ratio 3:1 - mix well and then place it in a platinum dish. Heat and fuse the contents of the dish and mix thoroughly by swirling. Let the flux cool and grind it with a porcelain mortar and pestles so that it passes a 10 mesh screen or finer.

2. Weigh a g of sample and then weigh 8 g of the ground flux, mix both well in a suitable platinum crucible.

3. Add known amount of ^{236}Pu for recovery.

4. Heat the mixture until dissolved (fused), then cool.

5. Place crucible in a plastic beaker containing 120 ml H_2O, 20 ml conc. H_2SO_4, 1 ml La carrier (20 mg/ml) and 1 ml H_2O_2 -when the fused mixture is released from crucible remove the crucible and rinse well with H_2O.

6. When reaction has stopped, stir with a teflon stirrer and add 10 ml conc. HF and let stand at least 1/2 hour.

7. Centrifuge the precipitate (LaF_3) in plastic tube.

8. Wash the precipitate with a solution of approximately 10 ml of 1 M HNO_3 and 1 M HF

9. Dissolve precipitate with a 1.2 ml of 2 M aluminum nitrate and 4 ml 2 M HNO_3, stir and heat lightly (in H_2O bath), allow approximately 5 minutes, then cool.

10. Add 1 ml 1 M hydroxylamine hydrochloride to solution and heat for 5 minutes, then cool.

11. Transfer the solution (which is 6.2 ml) into a small separatory funnel, preferably a 60 ml one.

12. Weigh out 0.276 g of $NaNO_2$ and dissolve it in 2 ml H_2O.

13. Add the above $NaNO_2$ solution to separatory funnel and mix well.

14. When reaction is complete add 8.2 ml 0.45 M TTA and then shake (extract) for 5 minutes.

15. Discard the aqueous phase.

16. Wash organic phase 2 times with 1 M HNO_3, shaking each time for 1-2 minutes.

17. Back extract Pu with equal volumes of 10 M HNO_3 twice, shaking for 1-2 minutes each time. Combine aqueous layers in a 100 ml beaker.

18. Evaporate to approximately 5 ml, then all 1 ml of conc. $HClO_3$ and 1 ml conc. H_2SO_4. Heat until fumes of $HClO_3$ have stopped. Sample is ready for plating.

19. Proceed to Step 28, Preparation and Plating of Sample of Procedure A.

(d) Determination of Thorium by Alpha Spectrometry

Application

This method is applicable to the analysis of various types of solubilized environmental samples.

Principle of Method

Lanthanum carrier is used to co-precipitate thorium as the fluoride with HF. Solution of the precipitate is accomplished by warming with HCl and H_3BO_3. The precipitation of lanthanum (Th) hydroxide removes the alkaline earths contamination. Solution of the hydroxide precipitate in concentrated HCl and the subsequent passage through an anion column removes several contaminating nuclides, since thorium is not sorbed on the anion column at any HCl concentration.

The effluent from the ion exchange column, which contains the Th, is evaporated to dryness. The residue is dissolved in 0.2M HNO_3 and the Th extracted with TTA to separate the Th from the La carrier.

The thorium is then plated on a 1" stainless steel disk. The total thorium activity is determined by measuring the alpha activity in an internal proportion counter. The isotopes of thorium present may be determined by alpha spectrometry.

Reagents

Lanthanum carrier - 12 mg La^{+++}/ml
Hydrofluoric acid, HF - 48%
Boric acid, H_3BO_3 - saturated solution
Hydrochloric acid, HCl - concentrated, 4M, 0.7M
Ammonium hydroxide, NH_4OH - concentrated
Anion exchange resin - Dowex AG 1X10, (200-400) mesh, chloride form, see Appendix at end of Method for preparation
Nitric acid, HNO_3 - concentrated, 0.2M, 4M
Perchloric acid, $HClO_4$ - 70%
2-Thenoyltrifluoroacetone, TTA - 10% w/v in benzene
Sulfuric acid, H_2SO_4 - concentrated, 0.7M
Methyl red or methyl orange

Special Apparatus

Electroplating apparatus
Electroplating cell
Stainless steel plating disks, 1 inch diameter
Internal proportion counter
Alpha scintillation spectrometer
Ion exchange columns

Procedure

1. To a suitable aliquot of a solubilized sample in a Teflon beaker add concentrated HCl until strongly acid. Add 1 ml of La^{+++} carrier.

2. Add 2 ml of hydrofluoric acid and mix thoroughly. Allow to stand for one hour for the precipitate to form completely. Decant. Discard the supernatant liquid. Transfer the precipitate to a 40 ml polycarbonate centrifuge tube. Centrifuge. Discard the supernatant liquid.

3. Dissolve the precipitate with 2 ml of a saturated solution of H_3BO_3 and 1 ml of concentrated HCl. Dilute to 30 ml with water.

4. Make the solution alkaline with concentrated NH_4OH. Centrifuge. Discard the supernatant liquid.

5. Dissolve the precipitate with concentrated HCl and sorb onto the Dowex anion column (see Appendix at end of Method column preparation).

6. Wash the column with 35 ml of concentrated HCl. Collect the effluent, which contains the thorium, in a 150 ml beaker.

7. Evaporate the effluent to dryness on a hot plate. Add 1 ml of concentrated HNO_3 and 1 ml of 70% $HClO_4$ and again take to dryness.

8. Dissolve the residue in approximately 10 ml of 0.2$\underline{M}$ HNO_3. Pour the solution into a 60 ml separatory funnel. Rinse the beaker with 1 ml of 0.2$\underline{M}$ HNO_3 and combine with the solution in the funnel.

9. Add 10 ml of 10% TTA solution to the separatory funnel and shake for 5 minutes. Draw off the aqueous layer into a clean 60 ml funnel.

10. Add 10 ml of 10% TTA solution to the funnel containing the aqueous layer and shake again for 5 minutes. Draw off and discard the aqueous layer.

11. Combine the organic phases into one of the funnels. Rinse the empty separatory funnel with 10 ml of 4$\underline{M}$ HNO_3 and add this to the funnel containing the TTA.

12. Shake the TTA-HNO_3 mixture for 5 minutes. Draw off the aqueous layer into a 150 ml beaker.

13. Add 10 ml of 4$\underline{M}$ HNO_3 to the organic phase in the separatory funnel. Shake again for 5 minutes. Draw off the aqueous layer and combine with the solution in the 150 ml beaker.

14. Add 1 ml of concentrated HNO_3 and 1 ml of 70% $HClO_4$ to the combined aqueous layers and take to dryness on a hot plate.

15. Add 1 ml of water to the electroplating cell.

16. To the beaker containing the sample add 7 drops of concentrated H_2SO_4 and 0.5 ml H_2O. Swirl beaker and transfer the thorium to the cell using a medicine dropper.

17. Wash beaker with 7 drops concentrated H_2SO_4 and 0.5 ml H_2O and transfer to the cell with a medicine dropper.

18. Wash beaker twice with approximately 0.5 ml H_2O and transfer to the cell with a dropper.

19. Add a drop of methyl orange or methyl red to the cell. Add fresh concentrated ammonia (or ammonia that has been stored under oil to keep free of CO_2) dropwise until there is a color change.

20. Back-titrate with 0.7$\underline{M}$ H_2SO_4 to the color change. Add two drops in excess. The volume should not be much above the stirrer. (~7ml)

21. Plate for one hour at approximately 450 milliamps, between 5 and 6 volts. Then add one dropperful of carbonate-free NH_4OH and plate for one minute. Pour solution into a beaker and save until plating is known to have been successful.

22. Disassemble the cell. Rinse plate in a beaker of H_2O containing some carbonate-free NH_4OH and ignite in a flame. If a stainless steel plate has been used, flame until the plated area just turns blue.

23. Count the plated sample in an internal proportion counter to obtain gross alpha counts. Then count in an alpha spectrometer.

Calculations

$$\text{total dpm} = \frac{\text{net cpm (IPC)}}{\text{efficiency}}$$

$$\text{pCi of Th isotope/aliquot taken} = \frac{\text{Counts in peak channels}}{(2.22)\ \text{total counts in all channels}} \times \text{total dpm}$$

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Appendix:

Preparation of anion column:

Dowex AG 1X10, Cl^- form, 200-400 mesh. A glass tube 16 mm in diameter and 17 cm-24 cm long is drawn out to a tip at one end. A glass wool plug is placed at this end. Add to the tube a water slurry of resin until it settles to a column height of 6.5 cm. Place a glass wool plug on top of the column. Wash the resin successively with 20 ml 12M HCl, 20 ml 4M HCl, 20 ml 0.7M HCl, and 20 ml of 12M HCl.

E. DATA HANDLING

INTRODUCTION

The quality of the data reported depends not only upon the care with which sampling and analysis are performed, but also upon the care with which calculations of the resulting data are performed, and upon the manner in which the data are presented in reports.[1] A key aspect of a quality assurance program is maintaining records that document every step of the process that leads to the data that ultimately are reported.

LABORATORY RECORDS

A measurement is useful if it is representative of the environmental material or parameter that is under study. Laboratory notes made during the analysis of a sample serve as a basis for judging the quality of the analysis, indicating whether any problems arose during the analytical procedures that might have adversely affected their outcome. For this reason, every effort should be made during sample preparation and analysis to record all aspects of the procedure that might reasonably be expected to affect the outcome of the analysis. As a general rule, every possibly relevant variable that is amenable to quantification should be recorded, even if only by a check mark on a form. These records may be needed not only by the person who is writing them, and only for the time period during which they are being written, but in many instances they may be needed by other persons and at some future time. It is important, therefore, that the notes be both legible and clear in meaning, so that others who read them will be able to reconstruct the events that are referred to.

It is often convenient to preserve electronically the data records from those instruments which provide digital electronic output signals. When this is done, great care must be taken to ensure that blocks of data are thoroughly annotated to identify the time and location and all other important circumstances concerning the measurements. Often the best way to do this is to organize blocks of data into files with distinctive file names, and to keep a logbook with the supporting information for each file. Alternatively, if electronic editing capabilities are available, comments can be entered into the data file itself.

DATA REDUCTION AND STORAGE

There are types of data, such as those related to possible legal actions involving the Government, that should be stored indefinitely. When data are stored electronically, backup files must be prepared to eliminate the possibility that the data may inadvertently be lost. When it begins to become evident that a medium on which data are stored, such as a particular type of magnetic disk or tape, is in danger of becoming obsolete, and there is thus a danger that the data will become irretrievable, the data should be copied to a better medium.

Whenever data are to be transferred or stored, an appropriate control procedure should be established to minimize the danger that human error will compromise the quality of the data. Data reduction is normally accomplished electronically using computer programs, both to avoid the drudgery of repeated "hand" calculations and to avoid calculation errors. For small numbers of unusual measurements or samples, however, hand calculations may be more efficient than writing a computer program of limited applicability. When unfamiliar calculations are performed, whether by hand or by using computer programs, care must be taken to be sure that the logic built into the calculation is correct. Calculations are often checked by performing them on data that lead to a known result.

DATA REPORTING

When data are reported, an estimate of their uncertainty must be given. Contained below are the guidelines and definitions of the terms that are useful for reporting the errors and uncertainties of data. The meaning of reported uncertainties must be indicated either by stating exactly what they represent or by describing how they were calculated, because a simple X ± Y statement may be interpreted in any one of a number of ways. The statement of uncertainty should include estimates of all significant sources of error involved, whether these result from the field measurement or sampling phase, the analysis phase or the data reduction phase, if they will affect the final result within the number of significant figures reported.

When data are reported, the reporting format must be commensurate with their expected use. Tables of data allow the full presentation of values and of their estimated uncertainties. Graphical presentation typically allows better visualization of the data. With both tabular and graphical presentations, it is important to assume that nothing will be immediately obvious to the reader, and the column headings or legends must include all information that is necessary in order to understand the data that are presented.

When data are presented, it is important to report only the appropriate number of significant figures. Usually the data should be carried to additional figures during preliminary calculations and then the final result should be rounded off to the proper number of significant figures. When the tables are printed by a computer, the format that is used may result in too many decimal places being reported for some samples. If this happens, the table should be edited to limit all data to the appropriate number of figures. One approach to determining the number of significant figures to be reported (Sanderson et al., 1980)(2) first determines the number of significant figures in the uncertainty, and rounds the reported value to the same decimal place as the uncertainty. In this approach, it is assumed that the rounded-off standard deviation reported should not differ from the calculated value of the standard deviation by more than 20%. For example, if the calculated standard deviation is 0.1635, it should be reported as 0.16, which differs from the original by over 22%. The value of the measurement is then rounded such that its last significant figure will be in the same decimal place as that of the error.

When data are presented in memoranda or reports, it is usually both possible and desirable to include a complete discussion of the quality assurance data that are pertinent to the measurements. The information to be presented may include, in addition to the uncertainties, various statistical tests and indications of analytical sensitivity, such as the lower limit of detection (LLD), the instrumental detection limit (IDL), the method detection limit (MDL), the limit of detection (LOD) Currie, 1988),(3)
the specificity (identification), and purity (absence of contaminant). The specificity and purity may be estimated from the resolution of a signal or from the goodness of fit to known quantities, such as energy, wavelength or rate of radioactive decay. Reporting the results for quality control samples, replicates and blank samples also provides important information about the quality of data. If a discussion of quality assurance cannot be included in a journal or symposium publication, normally a separate, supporting quality assurance report should be prepared.

Terms such as "below detection limits" should not be reported in place of the actual analytical results obtained. Reporting results as "less than" some minimum detectable level also results in some loss of statistical information, and may lead to erroneous interpretations. When low activities or concentrations are measured, the actual results obtained, including any negative values, normally should be reported along with the associated overall uncertainties and the measures of analytical sensitivity that are mentioned above. An interpretation of negative values can be included in the text. Waite et al. (1980)(4) and Gilbert Kinnison (1981)(5) have discussed techniques that may be followed in averaging data sets that contain "less than" values.

A number of terms have been used by the scientific community to describe data quality, but these terms are often given different meanings by different individuals. The definitions that follow are the preferred usage for the purposes of this document and have been taken from Croarkin (1985)(6), Taylor (1985, 1987)(7,8) and Taylor and Opperman (1986),(9).

Error - The difference between the true value and the measured value of a quantity or parameter.

Uncertainty - The range of values within which the true value is estimated to lie. It is a best estimate of possible inaccuracy due to both random and systematic errors.

Random Errors - Errors that vary in a nonreproducible way around the limiting mean. These errors can be treated statistically by use of the laws of probability.

Systematic Errors - Errors that are reproducible and tend to bias a result in one direction. Their causes can be assigned, at least in principle, and they can have both constant and variable components. Generally, these errors cannot be treated statistically.

A statement of uncertainty assigns credible limits to the reported value, stating to what extent that value may differ from its true value. The uncertainty of a measured value can be defined by a statistically determined confidence interval for the random error and by an estimate of the bounds for systematic error; they should be stated separately. When appropriate, they may also be combined into a single range to describe the overall uncertainty. Since there are a variety of methods suggested in the literature for combining random and systematic errors, the particular method used must be explicitly stated. One accepted practice (Croarkin, 1985)(6) is to combine in quadrature, systematic errors that are known to be independent, to add linearly systematic errors that may not be independent, and to combine systematic and random errors linearly. The confidence level chosen must be stated whenever a confidence interval is reported.

Systematic errors which can be determined by calculation or by experiment should be eliminated by an appropriate correction. Estimating the magnitude of some systematic errors may require scientific judgement on the part of the experimenter. All significant sources of error should be identified and reported.

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III RADIATION PROTECTION GUIDELINES

1. PHILOSOPHY OF RADIATION PROTECTION

The setting and execution of guidelines for radiation protection are based upon an underlying philosophy in which two factors are of prime importance. First is the assumption that radiation effects follow a linear, or non-threshold dose-response relationship. There is convincing evidence, particularly insofar as the genetic effects of radiation are concerned, that there exists a non-threshold phenomenon; and although positive proof is thus far lacking, it has been deemed prudent to adopt this more conservative hypothesis in setting protection standards for large numbers of people. According to the non-threshold concept, there is no radiation dose so small that it does not involve some degree of risk. The non-threshold relationship, therefore, implies that there is no radiation protection guideline, no matter how low, which can insure absolute safety to every individual in a large population receiving the guideline dosage. However, since the magnitude of the risk is proportional to the dose received, untoward effects would become manifest at very low dose levels only if extreme large numbers of exposed individuals were observed.

The second major factor to consider is that radiation, like many other developments of modern life (such as the automobile) confers great benefit upon both society and the individual along with its risk to health. Consideration of the extend of these benefits makes a certain degree of risk acceptable. Thus, a balance must be struck in each contemplated radiation usage, in which the benefit to be gained is weighed against the anticipated risk. If the benefit outweighs the risk, the radiation is utilized so that its maximum benefit will be realized while human exposure will be maintained at the minimum consistent with deriving these benefits. The overall public health philosophy, then, is to attain maximum advantage from the use of ionizing radiation while minimizing concomitant exposure, that is, eliminating wherever possible all unnecessary exposure to radiation.

The Radiation Protection Guide (RPG) may be defined as the radiation dose which should not be exceeded without careful consideration of the reasons for so doing. In light of the non-threshold phenomenon, every effort should be made to encourage the maintenance of radiation exposures as far below the guide as practicable. Methods of estimating guides are primarily based on past experience in which individuals have been exposed to radiation for various reasons and upon animal experiments which have contributed greatly to the study of the effects of radiation. From this combined knowledge and from an understanding of the relative biological damage produced by various types of radiation, protection guides for whole-body exposure and for various organs have been recommended. These guides, of course, represent doses far below those at which any effects have thus far been observed.

2. BASIS FOR RADIATION PROTECTION GUIDES

Establishment of "safe" levels of long-term radiation dose requires knowledge of the cause-effect relationship between radiation dose and biological damage. Such damage may appear many years after initial exposure and is usually indistinguishable from the normal diseases and impairments of man. Information accumulated on this subject is, therefore, difficult to evaluate and is often controversial. Nevertheless, observations involving man and animal life have resulted in the accumulation of significant data. These include:

A. Genetic Effects

1. Studies involving fruit flies and mice
2. Hiroshima and Nagasaki populations (inconclusive)

B. Incidence of Neoplasms

1. Bone tumors in radium dial painters
2. Leukemia in radiologists
3. Thyroid carcinoma in children irradiated in infancy
4. Various neoplastic diseases, including leukemia, in patients receiving therapeutic radiation for a variety of reasons
5. Leukemia in offspring of mothers irradiated during pregnancy.
6. Leukemia in survivors of Hiroshima and Nagasaki bombings

C. Life-Span Shortening

1. Studies with various animal species.
2. Radiologists vs. other physicians

Evaluations of these observations in terms of allowable radiation dose to man is an exceedingly complex task. The National Council on Radiation Protection and Measurements (NCRP) has accepted the task and over the past 30 years has established recommendations regarding maximum permissible doses. Because of the fragmentary evidence currently available, the NCRP points out that these levels are not necessarily completely safe limits but rather levels which should carry an acceptable risk.

In 1959, the Federal Radiation Council was formed to provide a Federal policy on human radiation exposure. This organization published its first report in May, 1960. Its recommendations were essentially the same as those of the NCRP and the International Commission of Radiological Protection (ICRP). The Council suggested, however, that the terms "Radiation Protection Guides" replace the former "Maximum Permissible Doses." The newer terminology recommended by the Federal Radiation Council is used throughout this section.

3. CURRENT RECOMMENDED DOSE LIMITS

Radiation Protection Guides vary depending on whether the whole body or only a portion thereof is exposed and whether radiation workers or the general public is involved.

A. Radiation Workers

1. ACCUMULATED DOSE (EXTERNAL SOURCES)

a. For external exposure to the whole body, the maximum average dose rate should not exceed 5 rems per year (0.05 Sv per year). The same dose rate applies to the head and trunk, active blood forming organs, the gonads, and the lenses of the eyes.

No occupational dose is allowed persons under 18 years of age. The accumulated dose at any subsequent age, therefore, should not exceed 5(N-18) rems (0.05(N-18) Sv), where N is the age of the worker in years.

b. Generally, however, where only a single portion of the body is exposed, Radiation Protection Guide average dose rates are higher. For example, it is recommended that the average dose rate to the hands and forearms and to the feet and ankles should not exceed 75 rems per year (0.75 Sv per year).

2. ACCUMULATED DOSE (INTERNAL SOURCES)

a. Certain elements, upon ingestion, inhalation, etc., deposit uniformly throughout the body. Radioisotopes of these elements deliver a dose to the whole body. The Radiation Protection Guide dose rate for whole body exposure from such radionuclides has the same limit as from external sources (i.e., 5 rems per year or 0.05 Sv per year). No occupational exposure is allowed persons under 18 years of age.

b. Other radionuclides tend to localize in one or more selected body organs. Based on the essentiality of the organ to proper body function, epidemiological experience with radium, and genetic considerations, other Radiation Protection Guides have been specified for selected critical body organs.

(1) For most individual organs, it is recommended that the maximum average dose rate not exceed 15 rems per year (0.15 Sv per year).

(2) For the thyroid and skin, the maximum average dose rate should not exceed 30 rems per year (0.30 Sv per year); for the gonads, the average dose rate should not exceed 5 rems per year (0.05 Sv per year).

(3) For compact bone, the maximum permissible dose rate is limited to the average dose rate which would result from a body burden of 0.1 microcurie (3.7×10^4 Bq) of radium. (This amount of radium would yield a dose rate of approximately 28 rems per year (0.28 Sv per year)).

B. General Population Groups

Recommended Radiation Protection Guides for the general population are considerably below those for radiation workers. One of the reasons for this stricter limitation on allowable dose is related to the possibility of genetic effects from radiation exposure. Because the general population contains all of the germ plasm controlling the genetic viability of the race, any mutational effects on this population would have a far greater total impact than exposure of that small segment of the population working with radiation.

Currently it is recommended that the yearly radiation exposure of individuals in general population (exclusive of natural background and the deliberate exposure of patients by dentists and doctors) should be held to one-tenth of the permissible occupational levels. Thus, for whole-body exposure of individuals in the general population, the radiation dose should not exceed 0.5 rem per year.

Exposure of the total population (this includes radiation workers and special groups, as well as the general population) presents a different problem. Here it is necessary to make assumptions concerning the relationship between the recommended Radiation Protection Guide of 0.5 rem per year (0.005 Sv per year) for individuals in the general population, and the average dose received by average total population groups. The Federal Radiation Council suggests the use of the arbitrary assumption that the majority of individuals do not vary from the average by a factor greater than three. Thus, they recommend the use of 0.17 rem (0.0017 Sv) as a Radiation Protection Guide for yearly whole-body exposure of average total population groups. This is in keeping with their recommendation that the average genetically significant dose to the total population not exceed 5 rems (0.05 Sv) up to age 30 (exclusive of natural background and purposeful exposure of patients by practioners of the healing arts).

Assessment of the amount of radiation dose received by a given individual or population group requires totalling the doses received from external and internal sources. External exposures may be measured with a sufficient degree of accuracy by direct roentgen-rate monitoring procedures. Internal doses are assessed in terms of the concentration of specific radionuclides in air, water, milk, and food which may be inhaled or ingested. To enable comparison of the radioactivity concentrations measured in these various environmental phases with Radiation Protection Guides , concentration values[1] which are related to the established RPG have been derived for many radionuclides.

[1] Concentration values are expressed in terms of Radioactivity Concentration Guides (RCG) or Maximum Permissible Concentrations (MPC).

C. Factors Influencing Radioactivity Concentration Guides

The Radioactivity Concentration Guide is the concentration of radioactivity in the environment which is determined to result in whole-body or organ doses equal to the RPG. In calculating RCG values for a given radionuclide, the following factors must be taken into consideration:

1. INITIAL BODY UPTAKE

Large fractions of some elements are absorbed when taken into the body. In the case of certain other elements, only small fractions are absorbed in passage through the gastrointestinal tract.

Therefore, the greater retention would increase the hazard from the first group as compared with the second, other factors being equal.

When radionuclides are inhaled, unless information specific to the radionuclide is available, it is assumed, in the case of soluble compounds, that 25 percent is retained in the lower respiratory tract. From here, the nuclides move into the blood stream and a portion of each is deposited in its critical tissue within a few days. Approximately 50 percent is held in the upper respiratory tract and is swallowed. In the case of insoluble compounds, it is assumed that 12 percent is retained in the lower respiratory tract, which is usually taken as a critical organ. The remainder is eliminated by exhalation and swallowing.

2. FRACTION RETAINED IN THE BODY

The rate of elimination from the blood and tissues of the body varies considerably for different elements or compounds. The time required for one-half of the original quantity of radioactive material to be removed from the body by biological processes is called the biological half-life.

Some materials in the blood stream are eliminated rapidly from the body whereas large fractions of others remain in the body organs. For example, radium, plutonium, and strontium are deposited in the bone where the rate of turnover is very slow; i.e.; the biological half-life is many years. Radioisotopes of these elements are much more hazardous than those of carbon, sodium, and sulfur which have biological half-lives of a few days or weeks. The principal biological methods of elimination of radionuclides from the body are the urine, feces, exhalation, and perspiration. Usually elimination is much more rapid before the radionuclide is translocated from the body to a more permanent area, such as the bone. This time is usually from a few days to a few weeks. After the initial period, the elimination rate becomes more nearly exponential, and the application of the term "biological half-life" has more meaning.

3. ORGAN OR TISSUE IN WHICH RETAINED[2]

a. Radiosensitivity - Certain body organs or tissues are more radiosensitive than others. For example, the lymphatic tissue and bone marrow are much more radiosensitive than muscle or nerve tissue. Therefore, in equal concentrations, an element like plutonium is more hazardous than uranium because the plutonium concentrates in the most sensitive part of the bone, whereas the uranium goes to other portions of the bone, to the kidneys, and to other relatively insensitive organs.

b. Size - For a given number of microcuries of a radionuclide in a critical organ, the smaller the organ the greater the concentration and the greater the dose delivered to the critical tissue. Iodine presents a much greater problem than sodium, since the iodine is very selectively absorbed in a much smaller organ, the thyroid gland, whereas sodium is rather uniformly distributed throughout the whole body. In many cases the radionuclide is deposited in a large organ but localized in a small portion of that organ, so that, in effect, the mass of the critical tissue may become very small.

c. Essentiality - Some organs are either not essential to the body function, or, when they are damaged or removed, special steps can be taken to supplement or compensate for their reduced function. For these reasons damage in some cases to the bone-marrow, kidneys, eyes, etc., may represent a greater hazard than equal tissue damage to the thyroid gland.

4. PHYSICAL PROPERTIES OF THE RADIONUCLIDE

a. Type and Energy of Radiation - The hazard is proportional to the relative biological effectiveness or quality factor of the radiation; e.g., alpha radiation is more damaging than beta radiation for a given energy of radiation. As the energy of the radiation is increased, the damage also increases.

b. Half-Life - The shorter the physical or radioactive half-life of a radionuclide, the less is the biological hazard. The radioactive half-life and the biological half-life may be combined into a single "effective" half-life by means of the formula:

$$T_{eff} = \frac{(T_r)\ x\ (T_b)}{T_r + T_b}$$

D. Calculations of Radioactivity Concentration Guides

As an example of the approach used in determining RCG values, the case of cesium-137 is illustrated.

1. The element cesium is distributed rather uniformly throughout the body. Therefore, an RPG for the radiation worker of five rems per year is selected for any radioisotope of this element.

2. Experimental and theoretical studies have shown that the presence in the body of 30 microcuries (1.11×10^6 Bq) of ^{137}Cs will yield a whole body dose rate of five rems per year. This quantity, 30 microcuries, is referred to as the "maximum permissible body burden" for ^{137}Cs.

[2] In certain instances, an organ of the body, such as the GI tract or lung, may receive a significant dose due to the mere presence of the radionuclide in material inhaled or ingested. Under these circumstances, the organ so exposed (even though not absorbing the radionuclide) may become the "critical organ."

3. If, however, 30 microcuries (1.11×10^{6} Bq) of ^{137}Cs were deposited within the body, they would not remain there long because of biological elimination and radioactive decay. Therefore, a certain amount could be ingested continually without exceeding the maximum permissible body burden.

4. A "standard man" consumes approximately 2200 milliliters of water per day. Approximately one-half of this is ingested during the working hours. If the daily intake of ^{137}Cs via drinking water is to be limited such that the "maximum permissible body burden" will not be exceeded (0-0.5 Sv) (and the average yearly radiation worker whole-body dose rate not exceed five rems), then the concentration of this nuclide in drinking water must not exceed 4×10^{-4} mCi/ml (14.8 Bq/ml), is the Radioactivity Concentration Guide (RCG) for ^{137}Cs in drinking water for consumption by radiation workers, based on an eight-hour per day, 40-hour work week for a radiation worker. .

In a similar manner, it can be shown that an average concentration of 6×10^{-8} Ci/ml soluble ^{137}Cs in air in the working environment would result in a worker receiving a whole-body dose rate of five rems per year.

E. Standardization of Calculations

When efforts were first made to establish permissible body burdens and Radioactivity Concentration Guide values for the various radionuclides, it became evident that it would be difficult, if not impossible, to compare suggested values from the different laboratories unless all used the same basic assumptions relative to the average man. Most of the calculations were based on the same fundamental physical assumptions relative to the energy schemes and radioactive half-life values, but each person making these calculations used his own preferred values for the mass and effective radius of the body organs, the rates of ingestion and inhalation, elimination rates, etc. This difficulty was recognized at several major conferences. As a consequence certain characteristics of the so-called "standard man" were agreed upon.

F. Graded System of Actions

It was felt by the Federal Radiation Council that a commitment to the single values represented by the Radioactivity Concentration Guides might prove arbitrary and inflexible, and not provide adequate guidance for taking assessment and control actions appropriate to the risk involved. Therefore, the Council's Report No. 2 introduced the concept of graded scales of RANGES of action in which average daily intake of a radionuclide by samples of exposed population groups are listed in three ranges. These ranges apply to certain selected radionuclides of health significance, namely, radium-226, strontium-89, strontium-90, and iodine-131. In general, the single radioactivity concentration guide equivalent corresponds to the upper limit of RANGE II. For intakes in RANGE I, the Council guideline recommends periodic, confirmatory surveillance as necessary; for RANGE II, quantitative surveillance and routine control is indicated; and for RANGE III, the Council advises evaluation and application of control measures as necessary.

G. Protective Action Guides

All of the foregoing guidelines are predicated upon the more or less constant release of low-level radioactivity into the environment from the routine uses of radiation, and assume continuous radionuclide intake by the population. Control, then, is based primarily at the source. There are cases, however, in which the contamination of the environment might be accidental or unforeseen, producing contamination which is transient and not likely to recur; these might include reactor incidents which result in relatively high but temporary local radioactivity levels, for example. In cases of this kind, the "contaminating event" would not occur on a regular basis, and control, or protective action, might be based upon limiting or changing the uptake of certain contaminated foods by the population. However, the impact of such measures upon the community which they are designed to benefit requires careful consideration by responsible authorities to insure that the benefit of the action taken is not outweighed by its other effects. To deal with this kind of situation, the Council, in its Reports No. 5 and No. 7, introduced the Protective Action Guide (PAG) concept. The PAG is defined as the projected absorbed dose to individuals in the general population which would warrant protective action following a contaminating event. It is assumed that the corresponding projected dose to a suitable sample of the exposed population would be one-third of the PAG. These guidelines have, thus far, been established for iodine-131, strontium-90, and cesium-137. It should be noted that a decision to take action to limit a community's intake of an important basic food item containing radionuclides involves a balancing of the health benefits to be attained against undesirable features of the protective action, such as disruption of dietary habits and nutritional needs. It follows, then, that control actions could be employed at dose levels above or below the PAG, depending upon the degree of total impact which the action has upon the community.

IV SUMMARY

Based on current information, Radiation Protection Guides for radiation dose have been recommended. These levels are subject to modification as more knowledge is gained. In view of their status and the possibility that any radiation dose may be damaging, it is well to recall the definition of the Radiation Protection Guide as that radiation dose which should not be exceeded without careful consideration of the reasons for doing so. Every effort should be made to encourage the maintenance of radiation doses as far below the Guide as practical. Table I summarizes the current recommendations of the ICRP and Federal Radiation Council. Table II are examples of the post-Chernobyl "action levels" as applied by various countries. Table III are examples of the Interim International Radionuclide Action Levels for Foods.

Table I summarizes the current recommendations of the ICRP and the Federal Radiation Council.

Table I. Radiation Protection Guides

Type of Exposure	**Condition**	**Dose[1](rem)[2]**
Radiation worker:	Accumulated dose	Five Times number of years beyond age 18
(a) Whole body, head, and trunk, Active blood-forming organs, gonads, or lens of eye	13 weeks	3
(b) Skin of whole body and thyroid	Year	30
	13 weeks	10
(c) Hands and forearms, feet and ankles	Year	75
	13 weeks	25
(d) Bone	Body burden	0.1 μCi of ^{226}Ra or its biological equivalent
(e) Other organs	Year	15
	13 weeks	5
Population:		
(a) Individual	Year	0.5 (whole body)
(b) Average	30 years	5 (gonads)

[1] Minor variations from other recommendations are not considered significant in light of present uncertainties

[2] Dose in Sieverts (Sv) is obtained by multiplyings by 10^{-2}

Table II. **Examples of post-Chernobyl "action levels" applied by different countries as of December 1986 for radionuclides in imported foods**

Country	Radionuclide	Food	Action Level[1] (Bq/kg or Bq/L
Brazil	^{134}Cs + ^{137}Cs	Milk powder	3700
		Other foods	600
Canada	^{131}I	Milk	10
		Dairy Products	40
		Other foods	70
	^{137}Cs	Milk	50
		Dairy Products	100
		Other foods	300
		Spices	3000
European Community Countries[2]	^{134}Cs + ^{137}Cs	Milk and infant foods	370
		Other foods	600
	^{131}I	Milk	500[3]
		Vegetables	350
People's Republic of China	^{131}I	Milk	1300
		Fruits and vegetables	270
		Cereals	340
		Beverages	130
	^{137}Cs	Milk	4600
		Fruits and vegetables	1000
		Cereals	1200
		Beverages	460
Poland	"Total Beta Activity"	Milk powder for infants and children to 6 years	1320
Sweden	^{131}I	All foods	2000
	^{137}Cs	All foods	300
USA	^{131}I	Infant foods	56
		Other foods	300
	^{134}Cs + ^{137}Cs	All foods	370

[1] Different terms used in different countries e.g. "Levels of concern" (USA), "Screening limits" (Canada), "Maximum permitted levels for import from third countries" (EC countries).

[2] Belgium, Denmark, France, Federal Republic of Germany, Greece, Ireland, Italy, Luxembourg, Netherlands, Portugal, Spain, United Kingdom.

[3] Recommended values by EC in May 1986.

Table III. Examples of Interim International Radionuclide Action Levels for Food (IRALF)

Radionuclide	Target Organ	Dose Level (mSv)	Dose Conversion Factor (Sv/Bq)	Radionuclide Intake (Bq corresponding) to the dose	Food Intake (kg)	IRALF (Bq/kg)
Sr-90						
-1st year	bone surface (infant)	50	1.9×10^{-6}	26,000	375	70
-following years		10	1.9×10^{-6}	5,200	375	20
I-131						
-1st year	thyroid (infant)	50	2.9×10^{-6}	17,000	40	400
Cs-134						
-1st year	whole body (adult)	5	2.0×10^{-8}	250,000	750	350
-following years		1	2.0×10^{-8}	50,000	750	50
Cs-137						
-1st year	whole body (adult)	5	1.4×10^{-8}	360,000	750	500
-following years		1	1.4×10^{-8}	71,000	750	100
Pu-239						
-1st year	bone surface (infant)	50	1.7×10^{-5}	3,000	375	10
-following years		10	1.7×10^{-5}	590	375	2

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IV GLOSSARY OF TERMS

INTRODUCTION

This Glossary of Terms has been included to assist the reader in understanding the abbreviations, symbols, and terms common to the radioassay procedures which appear in this manual. Many of the terms will be understood by anyone who has read the chapter on laboratory procedure included in most elementary quantitative analysis textbooks. Some will be familiar to those having basic knowledge of radiation physics. Others will be recognized as terms for which a traditional meaning has been extended to have a special meaning in certain contexts. A few, however, fall into the category of laboratory colloquialisms. Although these should be avoided in formal writing, they have been allowed to a limited extent, particularly in the step-by-step procedures.

All definitions have been restricted to the context in which the terms are used in this manual. The Glossary of Terms is neither a dictionary nor a complete glossary of terms. The rapid growth and development of scientific and technological knowledge precludes publication of any single complete glossary of current technical terms.

(1) ABBREVIATIONS AND SYMBOLS

A	activity, unless otherwise specified
AC	alternating current
α	alpha; alpha particles, energy of alpha particles, etc.
~, ≈	approximately
BP	boiling point
β	beta; as beta particles beta radiation, etc.
C	centigrade
Ci	curie(s)
cm	centimeter(s); 10^{-2} m
cm^2	square centimeter(s)
cph	count(s) per hour
cpm	count)s) per minute
d	day(s), in reference to half-life
dpm	disintegration(s) per minute
°	degree(s)
E	energy, unless otherwise stated
E_{max}	maximum energy
e.g.	*exempli gratia;* for example
et al.	*et alli;* and others
etc.	*et cetera;* and so forth
F	Fahrenheit
ft	foot (feet)
ft^3	cubic foot (feet)
g	gram(s)
GM	Geiger-Müller
γ	gamma; as gamma rays, gamma energy, etc.
h	hour(s), in reference to half-life
i.e	*id est;* that is
in	inch(es)
in^2	square inch(es), sq in
in^3	cubic inch(es), cu in
keV	kiloelectron volt(s); 10^3 electron volts
kg	kilogram(s); 10^3 grams
L or l	litre(s), liter(s)
λ	lambda; 1. decay constant 2. microliter; 10^{-3} milliliter
$\underline{M}$	molar, as applied to concentration of a substance in a solution
mA	milliampere(s); 10^{-3} ampere
max	maximum
meq	milliequivalent(s)
MeV	million electron volts

mg	milligram(s); 10^{-3} gram
min	minute(s)
ml	millilter(s); 10^{-3} liter
mm	millimeter(s); 10^{-3} m
μCi	microcurie(s); 10^{-6} curie
μg	microgram(s); 10^{-6} gram
-	minus
N	normal, as applied to concentration of a substance in a solution
nCi	nanocurie(s); 10^{-9} curie
pCi	picocurie(s); 10^{-12} curie
ppm	part(s) per million
/	per, devided by
%	percent, per hundred
+	plus
±	plus or minus, expressing a range
rpm	revolution(s) per minute
s, sec	second(s)
sp gr	specific gravity
σ	sigma; standard deviation
t	time, in hours days, etc.
$T_{1/2}$	half-life, equal to 0.693 devided by the decay constant
V	volt(s)
v/v	volume to volume; by volume
wt	weight
w/v	weight to volume; by weight
y	year(s), in reference to half-life

(2) DEFINITION OF TERMS AND LABORATORY PRACTICE

aliquot	*adj.*: 1. *Chem.* - fractional, as an aliquot portion of a chemical solution, implying a measure part. 2. *Math.* - contained an exact number of times in something else (*e.g.*, 5 is an aliquot part of 15) *n.*: an aliquot part; a measured portion of the whole
analytical balance	*n.*: a balance capable of weighing to 0.1 milligram
aspirate	*v.t.*: 1. to draw off or move fluids by suction; to draw off fluids with an aspirator. 2. to draw air through
background	*n.*: the sum of phenomena always present to affect physical apparatus or instrument response in the absence of a sample, above which a phenomenon must show itself in order to be measured. The term is applicable to radioactivity measurements (*e.g.*, counts or currents from cosmic rays), photometric measurements, etc., and is sometimes used in the sense of "blank determination".
blend	*v.t.*: to break down or mix to form a homogenous mass so that any portion of the mass is identical in composition, form, etc. to any other portion *n.*: a homogenous mass or mixture
carrier	*n.*: a substance in ponderable amount which will carry a trace substance with it through a chemical or physical process. If carriers are of the same element as the trace substance (*e.g.* 'Sr for ^{90}Sr), they are called *isotopic* carriers; if of a chemical similar element (*e.g.*, Ba for Ra), they are called *non-isotopic* carriers.
carrier recovery	*n.*: that fraction or portion of a carrier substance initially added which is finally recovered in the final product; esp., that which is finally recovered in a sample to be counted. Carrier recovery may be determined by weight, by flame spectrophotometry, or by other chemical or physical means.
centrifuge	*n.*: an apparatus for the separation of substances by centrifugal force *v.*: to separate by centrifugal force substances of different densities
chemical yield	*n.*: the quantity of a chemical product, such as a precipitate, obtained as a result of a chemical process expressed as a fraction or percentage of the amount theoretically obtainable [see yield]
count	*n.*: 1. the external indication of a device designed to enumerate ionizing events. Count may refer to a single detected event or to the total number of events registered in a given period of time; loosely, a disintegration, an ionizing event, or a voltage pulse. 2. (*Collog.*) a counting rate; the number of counts observed per unit time *v.t.*: to measure the radioactivity or a substance by enumerating the ionizing events it produces in a detector during a known time interval
count rate	*n.*: the number of ionizing events detected per unit time. *Background* count rate is the number of ionizing events registered by the detector system in unit time when no sample is present. *Net* count rate is the number of ionizing events registered by the detector per unit time in the presence of the sample excluding the background count rate.
daughter	*n.*: any radioactive or stable nuclide produced by the radioactive disintegration of a radionuclide, either directly or as the result of a series of transformations in a radioactive decay chain (^{90}Sr; ^{218}Po is a daughter of ^{222}Ra) *adj.*: designating a relationship in a radioactive decay series, as daughter product
decant	*v.t.*: to pour off gently; usually, to pour off the clear liquid above a settled or centrifuged residue

dry	*v.t.*: to remove the moisture from; to make devoid of moisture. Rigorously, *drying* means treatment in an oven at a temperature between 100° C and 110° C, but the term is extended to include the use of a hot plate or an infra-red lamp. *Drying* implies a more gentle treatment than igniting or baking. *adj.*: free of moisture; not wet or damp
dryness	*n.* the state or quality of being dry. *Take to* dryness implies the removal of liquid, but not prolonged baking.
equilibrate	[see radioactive equilibrium]
extract	*v.t.*: to obtain or withdraw from a composite substance by any chemical or mechanical process, as by distillation, dissolution, etc. *n.*: that which has been extracted, usually in a concentrated form
extraction	*n.*: the process of extracting or separating a certain substance from a mixture usually by means of solvents; exp., a process of separating or isolating a substance in solution by virtue of its varying degree of solubility in different solvents
gamma scan	*n.*: (*Colloq.*) a spectrum of gamma energies, usually in printed or plotted form, as that obtained from gamma pulse height analysis *v.: (Colloq.)* the process of obtaining a gamma spectrum; the process of examining a sample for gamma emitters by pulse height analysis or spectrometry
homogenate or homogenizing	*n.:* that which has been homogenized; the homogeneous mass resulting from blending
homogenized	*adj.:* uniformly blended; made homogenous; [see blend] - *Special:* with milk, to indicate treatment by a technical process using high temperature and pressure to break up and uniformly disperse fat globules.
ingrowth	*n.: (Colloq.)* the formation of a daughter product from its radioactive parent (*e.g.*, ^{90}Y from ^{90}Sr), usually applied to accumulation of the daughter product in a solution in which the radioactive parent has been isolated.
ion exchange	*n.*: a chemical process involving the reversible interchange of ions at a phase boundary, usually between a solution and a particular solid material, such as an ion-exchange resin *adj.*: ion-exchange
	The special terminology used in reference to this process is defined in the context of the following statement. < A solution is poured through an *ion-exchange column* which is a tube containing an *ion-exchange resin*. The solution that comes through is the *effluent* (general term). The material retained on the column (on the resin) is the *adsorbate*. The adsorbate is *eluted* from the resin by passage of a reagent solution, the *eluant*, through the column. The effluent from the process of *elution* is the *eluate*. The column (resin) is *washed* by pouring a reagent wash solution through it, usually to restore the resin to its original ionic state. >
isotope	*n.*: one of two or more species of atoms of the same element that have the same atomic number, occupy the same position in the periodic table, and are nearly identical in chemical behavior but differ in atomic mass or mass number and so behave differently in radioactive transformations. radioisotope: a radioactive isotope
nuclide	*n.*: one of two or more species of atoms of the same element that have the same atomic number, occupy the same position in the periodic table, and are nearly identical in chemical behavior but differ in atomic mass or mass number and so behave differently in radioactive transformations. radioisotope: a radioactive isotope
parent	*n.*: a radionuclide that disintegrates directly or through a series of transformations to yield a specified nuclide, the daughter; (*e.g.*, ^{238}U is the parent of all members of the uranium series including the stable end product ^{206}Pb) *syn.*: radioactive precursor

purity	*n.*: the state of being free from mixture or contact with contaminants, pollutants, or other substances which will interfere with a chemical process or end product
qualitative	*adj.*: 1. showing the nature or identity of any or all elements or materials in a compound or mixture without reference to the quantities present, not quantitative. 2. (*Colloq.*) improperly used to describe an analysis or analytical result in the sense of *approximate* or *rough*
quantitative	*adj.*: 1. determining or showing the quantity or amount of any or all elements in a given compound or solution or the components of a mixture. 2. complete, as quantitative transfer.
radioactive equilibrium	*n.*: the state attained among the members of a radioactive decay series or part of a series in which each parent has a longer half-life than its daughter, when the ratios between the activities of successive members remain constant, *i.e.*, the ratio of parent activity to daughter activity is independent of time. *Secular equilibrium* is approached, but never completely attained, when the half-life of the parent is so long that there is no appreciable change in the amount of parent during the time required for later products to attain equilibrium, so that equal numbers of atoms of all members of the series disintegrate in unit time, *i.e.*, all members of the series exhibit the same activity (*e.g.*, ^{226}Ra and its series through ^{214}Bi). *Transient equilibrium* will be reached if the half-life of the parent is so short that the quantity of parent decreases appreciably during the given time interval, so that all members of the series decrease in amount or activity exponentially with the half-life of the parent (*e.g.*, ^{222}Rn and its series through ^{214}Bi; ^{140}Ba to ^{140}La).
reagent grade	*adj.* or *n.*: conforming to American Chemical Society specifications for reagent chemicals as published in *Reagent Chemicals: American Chemical Society Specifications, 1960*, by the American Chemical Society Committee on Analytical Reagents
recovery	*n.*: the quantity of a product obtained as a result of a reaction or process or series of these, expressed as a fraction or percentage of that theoretically obtainable from the quantity of one of the chemicals or radioactive components initially present or added [see carrier recovery]
rinse	*v.*: to treat a container with water or other suitable liquid to facilitate a quantitative transfer, usually a mechanical action *n.*: the liquid agent used to facilitate a quantitative transfer
scavenger	*n.*: a substance added to remove impurities or to overcome the undesirable effects of one or more of the substances contained in a mixture (*e.g.*, Fe, Cd, and Zr are often used as scavengers to remove rare earths by precipitation.)
spectrometry	*n.*: the art or process of using a spectrometer, of measuring spectra, or of determining wavelengths or energies of the various radiation; implying measurement, as a spectrometer is a spectroscope fitted for measurements of the spectra observed with it; also, pulse height analysis
standard	*n.*: an established measure of volume, quantity, quality, or value; esp., a material of known composition which closely resembles in chemical and physical characteristics the sample to be analyzed and which is used for calibration *adj.*: having the accuracy or authority of a standard, serving as a gauge or model, as a standard weight
standard, radioactive	*n.*: a sample of radioactive material in which the number and type of radioactive atoms at a given reference time is known; a standard against which radiation measurement equipment can be calibrated

supernate	*n.*: a supernatant liquid or substance; esp., a liquid from which a precipitate has been thrown down; also, supernatant
surveillance	*n.*: a continuous scrutiny or inspection of an area, environment, population, etc.: - esp., a continuous program of detecting, measuring, recording, and evaluating the condition of an area, environment, or population with respect to its content of radioactive material or its exposure to radiation. *adj.*: pertaining to surveillance
transfer	*v.*: to convey from one place to another; esp., to remove a measured or known quantity of a sample from one container to another, as with a pipette *n.*: the act of making a transfer [see quantitative]
wash	*v.*: to cleanse, purify by immersing in or applying water or other liquid, often accompanied by agitation; - esp., to remove mother liquor from a precipitate by thorough mixing with water or other suitable liquid followed by separation by centrifugation or filtration. Treatment with several small portions of the liquid is much more effective than with one or two larger portions. Solution and reprecipitation is sometimes more effective in purifying than is washing. *n.*: the liquid used to cleanse or purify
water, deionized	*n.*: reagent water prepared by passage through mixed-bed ion-exchange resins in the H$^+$ and OH$^-$ forms
water, demineralized	*n.*: deionized water
water, distilled	*n.*: reagent water prepared by distillation
water, reagent	*n.*: water suitable for use in the analytical laboratory, prepared from tap water by distillation or deionization to contain not more than 5.0 parts per million of dissolved solids and to have an electrical conductivity of not more than 8 micro-mhos per centimeter at 25°C. Reagent water as defined above is suitable for the procedures of this manual; however, high purity reagent water should meet the more stringent American Chemical Society specifications for distilled water.
weightless	*adj.*: having such a small mass that it may be neglected, as for self-absorption corrections. A sample weighing less than 0.1 milligram per square centimeter may be considered weightless in determining its beta activity.
yield	*n.*: the quantity of a product obtained as a result of a reaction or process or series of these expressed as a fraction or percentage of that theoretically obtainable from the quantity of one of the chemical or radioactive components initially present or added [see carrier recovery]

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Radionuclides in food is a comprehensive introduction to radioactivity and radiation. Topics covered include natural and artificial radioactivity, radioactivity in the environment, metabolism of radiation in humans and animals and radionuclides in the food chain. The emphasis, however, is on analytical methodology that can be used reliably by the national food control authorities charged with protecting consumers from unsafe imported and locally produced food. The manual, written by an international expert, will be useful to all who are interested in problems related to radionuclide contamination of the environment and of the food supply in particular.

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